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I, Yukiko HAYASHI, the translator of the attached document,
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the attached document is a true English translation of Japanese
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Signed, this seventeenth day of January 2006

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[Title of the Invention]	External Additive for Electrophotographic Toner, Electrophotographic Toner, Electrophotographic Developer, and 10 Image Formation Method
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30	[Title of the Invention]	External Additive for Electrophotographic Toner, Electrophotographic Toner, Electrophotographic Developer, and Image Formation Method

[Scope of Claims]

[Claim 1] An external additive for electrophotographic toner, characterized in that the external additive is formed of an oxide fine particle comprising at least a silicon element, the oxide fine particle has a primary particle diameter R (average diameter) of 30 to 300 nm, a standard deviation δ of a particle size distribution of R is $R/4 \leq \delta \leq R$, and a shape of the external additive is substantially spherical with circularity SF1 and circularity SF2 of 100 to 130 and 100 to 125, respectively.

[Claim 2] The external additive for electrophotographic toner according to claim 1, characterized in that the oxide fine particle includes at least the silicon element and a metal element.

[Claim 3] The external additive for electrophotographic toner according to claims 1 and 2, characterized in that the oxide fine particle includes at least the silicon element and a titanium element.

[Claim 4] The external additive for electrophotographic toner according to claims 1 thorough 3, characterized in that an element composition of the oxide fine particle is evenly dispersed between a surface part and an inside part of the oxide fine particle.

[Claim 5] The external additive for electrophotographic toner according to claims 1 through 4, characterized in that the oxide fine particle is subjected to surface processing with at least an organosilicon compound.

[Claim 6] The external additive for electrophotographic toner according to claims 1 through 5, characterized in that the oxide fine particle is a hydrophobed oxide fine particle on which an $R^1_3SiO_{1/2}$ unit (R^1 is an identical or different monovalent hydrocarbon group each having 1 to 8 carbon atoms) is applied on a surface of the oxide fine particle.

[Claim 7] The external additive for electrophotographic toner according to claims 1 through 6, characterized in that the oxide fine particle is subjected to surface processing by at least

silicone oil and a liberation rate of the silicone oil is between 10 to 95 %.

[Claim 8] Electrophotographic toner including at least a binder resin and a colorant, and having a volume-average particle diameter of 2 to 7 μm , characterized in that at least the external additive for electrophotographic toner according to claims 1 through 7 is mixed in the toner.

[Claim 9] The electrophotographic toner, characterized in that the toner is mixed with the external additive for electrophotographic toner according to claims 1 through 7 and one or more kinds of external additive having a smaller average particle diameter of primary particle than the average particle diameter of primary particle of the external additive for electrophotographic toner.

[Claim 10] The electrophotographic toner according to claim 8 or 9, characterized in that the binder resin of the toner includes at least a polyol resin.

[Claim 11] The electrophotographic toner according to claim 8 or 9, characterized in that the binder resin of the toner includes at least a polyester resin.

[Claim 12] A two-component type developer, characterized by comprising a carrier formed of at least the toner according to any one of claims 8 through 11 and a magnetic particle.

[Claim 13] An image formation method, characterized by using the toner or the developer according to claims 8 through 12.

[Detailed Description of the Invention]

[0001]

[Technical Field of the Invention] The present invention relates to an external additive for electrophotographic toner, electrophotographic toner, an electrophotographic developer, an image formation method, and image forming apparatus.

[0002]

[Background Art]

A typical image forming process according to an

electrophotography method or an electrostatic printing method includes a development process, a transfer process, and a fixing process. In the development process, an electric latent image is formed by uniformly charging a photoconductive insulation layer, 5 subjecting the photoconductive insulation layer to an exposure, and dissipating electrical charges on an exposed portion of the insulation layer, and then the latent image is visualized by causing toner of charged fine powder to adhere to the latent image. In the transfer process, an obtained visible image is transferred onto 10 a transfer material such as transfer paper. In the fixing process, the obtained visible image is fixed by applying heat or pressure (normally, a heating roller is used). A two-component developer including a carrier and toner and a one-component developer not requiring the carrier (magnetic toner, non-magnetic toner) are 15 known as a developer for developing an electrostatic image formed on a face of a latent image carrier. A full-color image forming apparatus using a method of sequentially transferring a toner image of each color formed on a photoconductor to an intermediate transfer body to temporarily carry the transferred toner image and 20 re-transferring the carried toner image onto paper all at once is well known.

[0003]

The toner used in the electrophotography method or the electrostatic printing method, of which various kinds of 25 performances are required in each of the above-described processes, is mainly formed of a binder resin and a colorant, and additives such as a charging control agent and an offset prevention agent are added, if necessary. For instance, in the development process, in order to cause the toner to adhere to the electric latent image, 30 the toner and the binder resin for the toner is to maintain an amount of charge suitable for a copier or a printer, regardless of a surrounding environment such as temperature or humidity. Further, in the fixing process using a heating roller fixing method, the

toner is to have a satisfactory anti-offset characteristic such that the toner does not adhere to the heating roller which is normally heated to a temperature of about 100 to 230 °C, and is to have a satisfactory fixing characteristic with paper. Furthermore, a satisfactory blocking-resistant characteristic of the toner such that the toner does not induce blocking while being stored in the copier is also required.

[0004]

Further, in an electrophotographic field in recent years, high image quality is discussed from various angles. There is growing awareness that it is very effective to downsize a diameter of the toner and to form toner in a spherical shape. However, as the diameter of the toner is further downsized, a transfer characteristic decreases, and an image tends to have poor image quality. On the other hand, it is known that the transfer characteristic is improved by forming the toner shape spherical (Japanese Patent Application Laid-Open No. Hei9-258474). Under such circumstances, in a color copier field or a color printer field, image formation in higher speed is expected. In order to increase speed of the image formation, a "tandem method" is effective (Japanese Patent Application Laid-Open No. Hei5-341617, for instance). The tandem method is a method of obtaining a full color image on transfer paper by sequentially transferring and superimposing an image formed by an image forming unit on a single piece of paper which is conveyed to a transfer belt. Tandem method color image forming apparatus may use a wide variety of transfer paper, and has a distinguished characteristic that the full-color image with high quality may be obtained at high speed. Particularly, the characteristic that the full-color image may be obtained at high speed is specific to the tandem method color image forming apparatus, and other color image forming apparatus using other method does not have. On the other hand, an attempt to achieve high speed is made while keeping improvement of the image quality. In

order to achieve the high speed in an apparatus using the above-described method, it is necessary to reduce an amount of time required for paper to pass through a transfer part. Accordingly, it is necessary to increase transfer pressure in order to achieve a similar transfer capacity in a conventional apparatus. However, when the transfer pressure is increased, the transfer pressure causes the toner to aggregate at a time of transfer and the transfer may not be well performed. Accordingly, a problem of generating a hollow defect in a formed image occurs. To solve such a problem, a method of improving the image quality by specifying circularity, a particle diameter, specific gravity, and BET specific surface area of the toner, and specifying adhesive stress, and so forth, of the toner during compression at 1 kg/cm^2 to equal to or less than 6 g/cm^2 , is known (Japanese Patent Application Laid-Open No. 2000-3063, and so forth).

[0005]

However, when the adhesive stress under the compression at 1 kg/cm^2 is used, compression pressure is too low. Therefore, there are problems in image quality such as the transfer characteristic at when the transfer pressure increases by using such as an OHP film, heavy paper, and surface-coated paper, and the hollow defect in a character part. When the adhesion stress is further decreased, there is a problem of transfer dust, and so forth. Further, a method of improving a discharging characteristic of the toner by specifying an adhesion force of one particle of toner to equal to or less than $3.0 \text{ dyne/contact point}$ is also known (Japanese Patent Application Laid-Open No. 2000-352840). However, the adhesion force of the toner during the compression is not specified in the patent applicaiton. Accordingly, though the discharging characteristic is improved, it is ineffective in improving the image quality such as the transfer characteristic and the hollow defect in the character part.

[0006]

Further, for a purpose of improving developability and stability of the developability with time, a method of specifying an aggregation degree during the compression is also known (Japanese Patent No. 3002063). However, there is still a problem in the image quality such as the hollow defect in the character part only by specifying the aggregation degree during the compression, and it is difficult to fully improve the transfer characteristic and a transfer rate. Furthermore, an attempt to improve the hollow defect in the character part by specifying a product of the aggregation degree and a loose apparent density of the toner at 7 or less is also known (Japanese Patent Application Laid-Open No. 2000-267422). However, a physical behavior of the toner at the compression is not reflected and the attempt does not have enough effect in an intermediate transfer system, a development system with strong agitation, and so forth, in which the toner is subjected to higher stress. Further, an attempt to specify a ratio of the loose apparent density and a hardened apparent density of the toner to the loose apparent density/the hardened apparent density = 0.5 to 1.0, and to specify the aggregation degree to 25% or below (Japanese Patent Application Laid-Open No. 2000-352840) is also known. However, it is to be noted that the hardened apparent density used in the attempt is obtained from measuring a bulk density of the toner after tapping the toner 50 times. The hardened apparent density is similar to a physical behavior reflecting fluidity of the toner and may not reflect a factor which increasing the bulk density of the toner when a mechanical stress is applied to the toner. Similarly, the attempt does not have enough effect in the intermediate transfer system, the development system with the strong agitation, and so forth, in which the toner is subjected to the higher stress.

[0007]

On the other hand, for a purpose of improving the fluidity and a charging characteristic of the toner, and so forth, a method of using a mixture of a toner particle and inorganic powder of metal

oxide of various kinds, and so forth, is proposed. The mixture is referred to as an external additive. Further, for a purpose of improving hydrophobicity, the charging characteristic on a surface of the inorganic powder, and so forth, a method of processing the surface of the inorganic powder with a specific silane coupling agent, a titanate coupling agent, silicone oil, an organic acid, and so forth, or a method of coating the inorganic powder with a specific resin, is also known. As for the inorganic powder, powders of silicon dioxide (silica), titanium dioxide (titania), aluminum oxide, zinc oxide, magnesium oxide, cerium oxide, iron oxide, copper oxide, tin oxide, and so forth, are known.

[0008]

Particularly, a hydrophobic silica fine particle obtained from reaction of a fine particle of the silica or titanium oxide and an organosilicon compound such as dimethyldichlorosilane, hexamethyldisilazane, silicone oil, and then substituting a silanol base on the surface of a fine particle of the silica with an organic base, is used.

[0009]

Among such hydrophobing processing agents, the silicone oil is known to have sufficient hydrophobicity and have a low surface energy when included in the toner. Accordingly, the toner shows the distinguished transfer characteristic. Japanese Patent Publication No. Hei7-3600 and Japanese Patent No. 02568244 specify a hydrophobicity degree of the silica subjected to processing with the silicone oil. Further, Japanese Patent Application Laid-Open No. Hei 7-271087 and Japanese Patent Application Laid-Open Hei8-29598 specify an amount of the silicone oil to be added and a carbon content in an additive. A silicone oil content and the hydrophobicity degree described in the above patent document may satisfy hydrophobicity processing of inorganic particle, which is a base agent of the external additive, and secure stability of the charging characteristic of the developer under a high humidity

environment. However, no positive attempt is made to utilize the low surface energy of the silicone oil, which is an important characteristic of the silicone oil, to reduce adhesion of the developer to a member in contact with the developer such as a contact charging device, a developer carrier (sleeve), a doctor blade, a carrier, an electrostatic latent image carrier (photoconductor), and the intermediate transfer body. Particularly, background soiling due to a strong adhesion force of the developer to the photoconductor or a hollow defect after transfer (a portion in which the developer is not transferred) on a character part, a line part, and edge part of a dot part, and a center part in the image may not be improved only by adjusting the amount of the silicone oil to be added and the hydrophobicity degree. Further, the hollow defect is caused when a transfer member has a very rough surface and the transfer may not be performed on a concaved part, which is not improved in the above patent document likewise. An inorganic particle including a specific amount of the silicone oil as a liquid component is disclosed in Japanese Patent Application Laid-Open No. Hei11-212299. However, such a definition of the amount may not satisfy the above-described characteristics.

[0010]

Further, even and stable charging is required of the electrophotographic toner. When the charging is not satisfactorily even or stable, the background soiling and uneven toner density may occur, resulting in deterioration in the image quality. Further, along with downsizing of image forming apparatus, a development mechanism is downsized. Therefore, toner charge rise becomes more important factor to obtain high image quality. In order to improve the above-described situation, various proposals are made so far. Take an example of a proposal for improving the charging characteristic by using an additive for the electrophotographic toner. Japanese Patent Application Laid-Open No. Hei3-294864 discloses a nonmagnetic one-component developer

including inorganic powder body subjected to processing with the silicone oil. Japanese Patent Application Laid-Open No. Hei4-204665 discloses a one-component base magnetic developer with 3 to 30 % covering rate of the additive relative to the toner.

5 Japanese Patent Application Laid-Open No. Hei4-335357 discloses toner and an electrostatic developer, wherein a fine particle having a BET specific surface area of 5 m²/g to 100 m²/g are fixed on a surface of the toner, and a particle having a specific surface area of 1.2 time or more of the specific surface area of the fine particle

10 fixed on the toner are included in the electrostatic developer. Japanese Patent Application Laid-Open No. Hei7-43930 discloses a developer using nonmagnetic one-component toner including a hydrophobic silica fine particle and specific hydrophobic titanium oxide. Further, Japanese Patent Application Laid-Open No.

15 Hei8-202071 discloses a developer including an additive for toner composed of organic/inorganic matter composite particle including an organic polymer skeleton and a polysiloxane skeleton.

However, with the above-described proposals, sufficient evenness in the charging may not yet be obtained or an amount of the toner

20 charge rise may not be sufficient. Further, environmental stability relative to the amount of toner charge, particularly stability relative to the humidity, are not always satisfactory. Particularly, when the additive with improved hydrophobicity as a result of a surface processing of a regular oxide particle is

25 used, which may be seen in many proposals, desired charging stability may be obtained in an early stage. However, there is a problem such as occurrence of deterioration of the toner related to varying composition of the additive with time during operation, and so forth. Further, with a composite particle synthesized by

30 using a liquid phase method as in the Japanese Patent Application Laid-Open No. Hei8-202071, satisfying hydrophobicity may not be obtained or the hydrophobicity may change with time due to influence of a mediating substance remaining inside the particle.

[0011]

On the other hand, an attempt is made to prevent color misregistration and contamination, and to stabilize an image density and the transfer characteristic for a long time by adding
5 a large-diameter inorganic fine particle having an average particle diameter of 50 to 120 nm in the toner (Japanese Patent No. 3148950). However, no positive effect is achieved on charge rise after printing a number of sheets, the background soiling on a non-printing part in a high temperature/humidity environment or
10 in a low temperature/humidity environment, and so forth. Further, the additive for the electrophotographic toner formed of an oxide fine particle obtained by an oxidizing solid solution fine particle including two or more kinds of elements, characterized in that a minimum value of difference in first ionization potential
15 between the elements included in the solid solution fine particle is 20 to 4.20 eV and a maximum value of the first ionization potential of the element included in the solid solution fine particle is equal to or less than 9.00 eV, is also known. However, a particle diameter and a shape of the inorganic fine particle are not fully discussed.
20 Accordingly, the fluidity and the transfer characteristic of the toner are not satisfactory, and the toner may not be sufficiently agitated upon development only by specifying the ionization potential.

[0012]

25 On the other hand, in terms of required characteristics of the binder resins for use in the toner, that is, transparency, an insulating characteristic, water resistance, fluidity (as a powder body), mechanical strength, glossiness, thermal plasticity, grindability, and so forth, a polystyrene, a styrene-acrylic
30 copolymer, a polyester resin, and an epoxy resin are generally used as the binder resin. Among them, a styrenic resin is commonly used for its satisfactory grindability, water resistance, and fluidity. However, when a copy obtained by using toner including the styrenic

resin is stored in a document holder made of a vinyl chloride resin sheet, an image face of the copy is left closely contacting the sheet. Therefore plasticizer included in the sheet, that is, the vinyl chloride resin, transfers and plasticizes on a fixed toner image and causes the fixed toner image to adhere on a sheet side. As a result, when the copy is released from the sheet, a part or whole of the toner image is peeled off from the copy, causing a defect such that the sheet is smeared. Such a defect may be found in the toner including the polyester resin.

[0013]

To prevent the toner from transferring on the vinyl chloride resin sheet as described above, Japanese Patent Application Laid-Open No. Sho60-263951 and Japanese Patent Application Laid-Open No. Sho61-24025 disclose a proposal of blending an epoxy resin which may not be plasticized by the plasticizer for the vinyl chloride resin in the styrenic resin or the polyester resin.

[0014]

However, when such a blended resin is particularly used in color toner, an off-set characteristic, curling of a fixed image, glossiness (a color toner image with insufficient glossiness may be seen as an image with poor image quality), a staining characteristic, a transparency characteristic, and a coloring characteristic may become a problem due to a non-phase solubility characteristic between different resins. Not all the above-described problems may not be solved by the conventional epoxy resin or an acetylated modified epoxy resin proposed in Japanese Patent Application Laid-Open No. Sho61-235852.

[0015]

The above-described problems may be solved by a single use of the epoxy resin. However, a new problem relating to reactivity of the epoxy resin with amine may occur. By forming a cross linked structure as a result of a reaction between an epoxy group and a curing agent, the epoxy resin is generally used as a cured resin

having a satisfactory mechanical strength and chemical resistance. The curing agent is roughly classified into an amine based curing agent and an organic acid anhydride curing agent. Naturally, the epoxy resin used as the electrophotographic toner is used as a heat plasticity resin. However, some dyeing pigments and the charging control agent kneaded with the resin to be used as toner are amine-based, accordingly a cross linking reaction may occur during kneading. Accordingly, the amine-based dyeing pigment and the charging control agent may not be used as the toner. In addition, a chemical activity of the epoxy group may potentially induce biochemical toxicity such as skin irritation. Sufficient attention must be given to existence of the epoxy group.

[0016]

Further, since the epoxy group shows hydrophilicity and significantly absorbs water in the high temperature/humidity environment, charge decrease, background soiling, improper cleaning, and so forth, are caused. Furthermore, charging stability in the epoxy resin is another problem.

[0017]

Regular toner is composed of the binder resin, the colorant, the charging control agent, and so forth. Various kinds of dyeing pigments are known as the colorants. Some of the colorants show a charge control characteristic and thereby acts as two agents, that is, the colorant and the charging control agent. It is popularity practiced to use the epoxy resin as the binder resin to compose toner having a composition as described above. However, there is a problem in a dispersion characteristic in the dyeing pigment, the charging control agent, and so forth. Generally, the binder resin are kneaded with the dyeing pigment, the charge control agent, and so forth, by a heat roll mill, and it is necessary to evenly disperse the dyeing pigment, the charge control agent, and so forth, in the binder resin. However, it is difficult to achieve thorough dispersion. Insufficient dispersion of the dyeing

pigments functioning as the colorant results in insufficient coloring and a low coloring degree. Further, insufficient dispersion of the charging control agent, and so forth, results in an uneven charge distribution, which causes various defects such as improper charging, background soiling, scattering, insufficient ID, fuzzing, and improper cleaning. Further, Japanese Patent Application Laid-Open No. Sho61-219051 discloses toner using an ester-modified epoxy resin, modified with ϵ -caprolactone, as the binder resin. Although vinyl chloride-resistance and the fluidity may be improved, since the epoxy resin used here is modified from 15 to 90 % by weight, there is a defect such that softening point is excessively lowered and glossiness is excessively high.

[0018]

Japanese Patent Application Laid-Open No. Sho52-86334 discloses an epoxy resin having a positive charge characteristic obtained by causing aliphatic primary or secondary amine to react with a terminal epoxy group of an existing epoxy resin. However, as described above, the epoxy group induces the cross linking reaction with the amine and there may be a case that the resulting resin may not be used as toner. Further, Japanese Patent Application Laid-Open No. Sho52-156632 discloses an attempt of causing any one or both of the terminal epoxy group of the epoxy resin to react to an alcohol, a phenol, a Grignard reagent, an organic acid sodium acetylide, an alkyl chloride, and so forth. However, if there is a residual epoxy group, it causes problems, as described above, in the reactivity with amines, the toxicity, the hydrophilicity, and so forth. Further, some of the above-described reaction products are hydrophilic, or affect the charging or grindability when composing toner, and are not always effective to the present invention.

[0019]

Further, Japanese Patent Application Laid-Open No. Hei1-267560 discloses a modified epoxy resin obtained by causing both of the

terminal epoxy groups of the epoxy resin to react to a monovalent compound having active hydrogen and then esterifying a reaction product with a monocarboxylic acid, or an ester derivative and lactones of the monocarboxylic acid, in which the reactivity, the toxicity and the hydrophilicity of the epoxy resin are solved. However, curling in fixing is not sufficiently improved.

[0020]

Further, a solvent such as xylene is generally used when synthesizing the epoxy resin or a polyol resin (for instance, Japanese Patent Application Laid-Open No. Hei11-189646). However, there is a problem that the solvents and unreacted residual monomers such as bisphenol A, and so forth, inevitably exist in the produced resin and consequently an amount of the residual solvents is large in toner using the resins.

[0021]

On the other hand, as to a production method of the toner, a typical method disclosed in Japanese Patent Application Laid-Open No. Hei1-304467 is generally used. In the method, materials are mixed all at once. Then a resulting mixture is subjected to heating, melting, and dispersing by a kneading device to form a homogenous composition. Further, the resulting mixture is subjected to cooling, pulverizing, and classification to produce toner having a volume-average particle diameter of 6 μm to 10 μm . Particularly, electrophotographic color toner for use in formation of a color image generally includes various chromatic dyes or pigments dispersed in the binder resin. In this case, performance required of the toner to be used is more strict than in a case when the toner is used to obtain a black image. That is, in addition to mechanical and electrical stability to external factors such as an impact and humidity, appropriate color development (colorability) and optical transparency when used in an over head projector (OHP) sheet are required of the toner. Toner using a dye as a colorant is described in Japanese Patent Application Laid-Open No. Sho57-130043 or

Japanese Patent Application Laid-Open No. Sho57-130044, for instance.

[0022]

When the dye is used in the colorant, a sharp color image with distinguished transparency and good coloring may be formed, however, it is less resistant to light. Accordingly, when the obtained color image is left under direct radiation, there is a problem of discoloration and color deterioration.

[0023]

Image forming apparatus according an intermediate transfer method is known. In the image forming apparatus, a plurality of visible color developed images sequentially formed on an image carrier are subjected to a primary transfer by being sequentially superimposed on an intermediate transfer body moving endlessly. Then a primary transfer image (toner image) on the intermediate transfer body is then subjected to a secondary transfer by transferring on a transfer material all at once. The image forming apparatus using the intermediate transfer method is advantageous in downsizing and that there is little restriction on a kind of transfer material on which a visible image is finally transferred. Therefore, the above-mentioned image forming apparatus tends to be used as color image forming apparatus. In such image forming apparatus, so-called a worm-eaten shape part (hollow defect of a character) sometimes occurs in an image on the transfer material such as transfer paper, or a final image medium, due to local transfer omission in a transferred toner image constituting the color developed image in the primary and the secondary transfer. In a solid image, an image having a worm-eaten part form transfer omission with a certain area. Further, in a line image, the transfer omission occurs forming a broken line image.

[0024]

Such an abnormal image easily occurs when forming a four-color image. This is because in addition to a thick toner layer, since

the primary transfer is repeated at maximum four times, a strong mechanical adhesion force, or a non-Coulomb force, (van der Waals force and other force except electrostatic force) is generated due to a contact pressure between a surface of the image carrier and the toner, or between a surface of the intermediate transfer body and the toner. In addition, a filming phenomenon, in which the toner adheres on the surface of the intermediate transfer body forming a film, occurs in a process of repeatedly performing an image formation process, and thereby the adhesion force between the surface of the intermediate transfer body and the toner increases. This is considered to be another reason of the abnormal image.

[0025]

To avoid such a worm-eaten shape image from occurring, techniques of reducing the adhesion force between the image carrier or the intermediate transfer body and the toner by applying lubricant on the surface of the image carrier or the intermediate transfer body, or reducing the adhesion force of the toner by an external additive, are already put to practical use in commercially available apparatus. However, in the techniques, the adhesive force among toner particles, tensile rupture strength, and so forth, when a transfer contact pressure generated during full four-color image formation or transfer at a high speed is increased, is not considered. Particularly, there is a problem in image stability when transfer is performed on the heavy paper, the surface-coated paper, the OHP film, and so forth.

[0026]

In addition, Japanese Patent Application Laid-Open No. Hei8-211755 discloses a method of improving the transfer characteristic and preventing the abnormal image with a worm-eaten-like hollow defect from occurring by adjusting a relative balance between the adhesive force of the toner on the image carrier and the adhesive force of the toner on the intermediate transfer body. However, the adhesion force of the toner in the case

is obtained by a centrifugal method when the toner is in a powder state, and the toner has different physical property from the physical property of the toner under increased transfer contact pressure.

5 [0027]

Toner after production faces a severe condition such as a high temperature/humidity environment or a low temperature/humidity environment during storage or transportation. There is a demand for toner having high storage stability with no or little
10 deterioration in a charging characteristic, fluidity, a transfer characteristic, and a fixing characteristic without aggregation among toner particles even after the toner is stored under such severe conditions. However, no effective means for the demand is not achieved so far.

15 [0028]

[Problems to be Solved]

An object of the present invention is to provide:

1. an external additive for electrophotography, toner, a developer, and an image formation method, in which the external additive
20 may not be buried in the toner and may satisfactorily exert functions as a fluidizing and a charge auxiliary agent even when the toner is subjected to agitation and charging after being stored under a high temperature/humidity environment, and capable of providing stable image quality by preventing charge
25 from abnormally increasing even after the toner is stored in a low temperature/humidity environment;
2. an external additive for electrophotography, toner, a developer, and an image formation method capable of forming a high-quality
30 material and with excellent transfer characteristic, developability, and a fixing characteristic, wherein an aggregation characteristic after the toner is subjected to stress during transfer/compression or in development apparatus,

and an adhesion force among toner particles are appropriately controlled;

3. image forming apparatus and an image formation method for forming an image with less background soiling (fog) with toner having excellent charging stability with less weakly and reversely charged toner in a high temperature/humidity environment or in a low temperature/humidity environment, and with less toner dispersion into the apparatus;

4. an image formation method with high durability and good maintainability as an image forming system;

5. an image formation method in which toner has an excellent supply characteristic and a charge rise characteristic with sufficient fluidity when the toner is not under compression, as well as an excellent transfer characteristic of the toner under compression;

6. image forming apparatus and an image formation method with a well-balanced fixing characteristic and an anti-offset characteristic capable of stably charging toner and a developer under any environment, equally printing an image at any speed from low to high, and forming an image without lowering image density when continuously outputting images;

7. an image formation method capable of forming an image with excellent color reproducibility, color sharpness, and color transparency, and with stable glossiness without surface irregularity under good transfer status of toner;

8. an image formation method with excellent environmental stability and environmental storage stability;

9. an image formation method in which a toner image may not be transferred onto a vinyl chloride resin sheet even when a surface of a fixed image is closely contacting the sheet;

10. an image formation method in which a fixed image may not virtually curl ; and

[0029]

11. an image formation method capable of preventing abnormal image such as an image with a hollow defect, dust deposition, and poor reproducibility of a thin line from occurring by using image forming apparatus according to a method of subjecting a toner image formed on an electrostatic image carrier to a primary transfer on an intermediate transfer body and the toner image is then subjected to a secondary transfer on a transfer material, and/or image forming apparatus according to a tandem method capable of outputting an image at a high speed.

[0030]

[Means for Solving the Problems]

To solve the problems, the inventor of the present invention keenly examined the problems. As a result, it is found that by using an external additive for electrophotographic toner characterized in that the external additive is substantially spherical and formed of an oxide fine particle including at least a silicon element, a primary particle diameter R of the oxide fine particle is within a range of 30 to 300 nm, a standard deviation δ of a particle size distribution of R is $R/4 \leq \delta \leq R$, and a circularity SF1 and a circularity SF2 are from a range of 100 to 130 and a range of 100 to 125, respectively, sufficient fluidity may be achieved. Further, by using the external additive, even after toner is stored under a high temperature/humidity environment, the external additive is not buried in the toner and may sufficiently exhibit functions as a fluidizing and a charge auxiliary agent, on the other hand, even after the toner is stored under a low temperature/humidity environment, an image with stable quality may be provided by preventing charge from abnormally increasing, furthermore, by using the external additive, an image with high quality with an excellent transfer characteristic and developability may be formed, wherein an aggregation characteristic of the toner during transfer and compression, and adhesion force among toner particles after the toner is subjected to stress in development apparatus are

appropriately controlled.

[0031]

Although a mechanism of the external additive is presently under study, following assumption may be made from some analysis data.

5 By specifying the primary particle diameter of the oxide fine particle within a range of 30 to 300 nm, the oxide fine particle fully exert a spacer effect of preventing aggregation among the toner particles and may achieve an effect of preventing additives from being buried in the toner when the toner is stored at a high

10 temperature is deteriorated as a result of intensive agitation. Further, by specifying the standard deviation δ of the particle size distribution R within $R/4 \leq \delta \leq R$, the oxide fine particle having a large particle diameter, a medium particle diameter, and a small diameter are included in the oxide fine particle in

15 appropriate proportion, wherein the particle having a small diameter secures the fluidity while the particle having a medium or a large diameter effectively exert the spacer effect. It is found that by specifying the standard deviation δ of the particle size distribution R within $R/4 \leq \delta \leq R$, the particle functions more

20 effectively than simply mixing each particle having a large particle diameter, a medium particle diameter, and a small particle diameter. In addition, by specifying the circularity SF1 and the SF2 of the oxide fine particle as 130 or less and 125 or less, respectively, to be substantially spherical, the fluidity of the toner and

25 affinity of the oxide fine particle for the toner increase. As a result, the oxide fine particle are prevented from being released from the toner, exerting an inherent function as an external additive.

[0032]

30 In addition, by using an external additive for electrophotographic toner characterized in that the oxide fine particle in the external additive includes at least the silicon element and a metal element, the external additive may sufficiently

achieve functions as the charge auxiliary agent, an agent for imparting appropriate electric resistance, and the fluidizing for the toner. Further, the metal element (dopant compound) may adjust a charging level and a resistance level of the toner. Oxide fine particle having various characteristic such as a dielectric characteristic and a resistance characteristic may be easily produced by controlling a composition of an original solid solution fine particle and a degree of oxidization of the oxide fine particle. By using the oxide fine particle, the charging characteristic of the electrophotographic toner is easily controlled within a desired range.

[0033]

In addition, to prevent a residual image generated during development, it is desirable to set electrical resistance on a developer carrier relatively low so that a residual electrical charge immediately leaks in the developer carrier. However, in such a developer carrier, an electrical charge to be held by the toner also leaks sometimes. A fine particle of the additives of the electrophotographic toner according to the present invention is stable and prevents the electrical charge from leaking. Accordingly, by using the silicon element and the metal element, an above-described problem may be solved.

[0034]

In addition, the charging level and the resistance level may be appropriately controlled by the oxide fine particle including at least the silicon element. Accordingly, environmental stability of charging when composing toner may be improved. The oxide fine particle may include a general substance in addition to SiO_2 , as long as the substance is according to the present invention. Examples of the substances include MgO , CaO , BaO , Al_2O_3 , TiO_2 , SnO_2 , combinations of two or more of the substances, and so forth. Among the substances, by particularly including at least a silicon oxide and a titanium oxide in the oxide fine particle, the toner particle

may be provided with the excellent fluidity, the charging characteristic, and durability during intensive agitation of the toner.

[0035]

- 5 By the composition of the oxide fine particle evenly dispersed between a surface part and an inside part of the oxide fine particle, the external additives for the electrophotographic toner having excellent stability with less variable dielectric characteristic/resistance characteristic may be achieved.
- 10 By giving surface processing by at least an organosilicon compound, the oxide fine particle may have more stable charging characteristic. The oxide fine particle obtained by a method according to the present invention may sometimes become an unsaturated oxide under some conditions for oxidizing the solid solution fine particle. In this
- 15 case, a characteristic of the additive may vary due to a development of oxidization with time. To prevent the variation with time, a reactive portion of the fine particle of the additive may be preferably inactivated. Surface processing by an organosilicon compound surface processing agent and/or an organotitanium compound
- 20 surface processing agent are particularly preferable. The surface processing according to hydrophobing processing is furthermore preferable.

[0036]

- To further improve the environmental stability of charging, the
- 25 oxide fine particle is preferably a hydrophobed spherical silica fine particle of which an $R^1_3SiO_{1/2}$ unit is used on a surface of the oxide fine particle. R^1 is an identical or different monovalent hydrocarbon group each having 1 to 8 carbon atoms. Examples of such monovalent hydrocarbon groups are a methyl group, an ethyl group,
- 30 a propyl group, a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a cyclohexyl group, a phenyl group, a vinyl group, an allyl group, and so forth. Among them, the methyl group is particularly preferred.

[0037]

Further, by subjecting the oxide fine particle to the surface processing at least by silicone oil, wherein a liberation rate of the silicone oil is from 10 to 95 %, the silicone oil, of which an amount is appropriately controlled, covers a surface of the toner. Accordingly, environmental storage stability may be improved. If the liberation rate of the silicone oil is less than 10%, a characteristic of the silicone oil may not be fully achieved. In addition, it is not desirable if the liberation rate is more than 95% because the silicone oil may adhere on an electrostatic image carrier and cause filming, and so forth. Further, the fluidity of the undesirably decreases.

[0038]

In addition, by mixing the oxide fine particle in the electrophotographic toner having a small particle diameter such as a volume-average particle diameter of 2 to 7 μm and including at least a binder resin a colorant, the toner having a small particle diameter may be prevented from being easily aggregated and the fluidity may be prevented from lowering. Accordingly, the charging stability and environmental storage stability may be improved.

[0039]

In addition to the oxide fine particle, by mixing one or more kinds of external additive having a small average particle diameter of a primary particle, the fluidity which may not be fully achieved by a large-diameter oxide fine particle having a diameter within a range of 30 to 150 nm alone, may further be improved. Further, covering rate of the external additive relative to the toner and affinity of the external additive between the external additives may be improved. Accordingly, favorable adhesion status of the external additive is achieved.

[0040]

By at least including a polyol resin in a binder resin of the toher, the toner may have excellent charge matching with the oxide

fine particle, sufficient compressive strength, tensile rupture strength, environmental stability, and a stable fixing characteristic. Further, by at least including the polyol resin having an epoxy resin moiety and a polyoxyalkylene moiety in the binder resin of the toner, the environmental related stability, the stable fixing characteristic may be achieved, and a toner image may be prevented from transferring onto a vinyl chloride resin sheet when a surface of a fixed image of a copy is closely contacting the sheet. Particularly, when the binder resin is used in color toner, it is effective for color reproducibility, stable glossiness, and in preventing the fixed image of copy from curling.

[0041]

In addition, by including at least a polyester resin moiety in the binder resin of the toner, the toner may achieve a well-balanced stretching characteristic and an adhesion characteristic as well as a compressive strength. Accordingly, a further stable transfer characteristic, developability, and a fixing characteristic are achieved.

[0042]

In addition, by using a two-component development system characterized by including at least a carrier composed of the toner and a magnetic particle, the developing system may achieve well-balanced adhesion to the carrier, less stress variation, and a sufficient bulk density as a developer, and a development characteristic excellent in a charge rise characteristic and environmental stability of charging. Further, a development system may satisfactorily control a toner density by a bulk density sensor, and so forth.

[0043]

An image forming apparatus for forming a toner image by developing an electrostatic image on an electrostatic image carrier by an electrostatic image developer, and electrostatically transfer the toner image on a transfer material by contacting a transfer means

on a surface of the electrostatic image carrier via the transfer material, characterized by using a two-component developer composed of a carrier including a magnetic particle and the electrophotographic toner, is achieved.

5 [0044]

According to the present invention, following (1) through (14) are provided.

(1) An external additive for electrophotographic toner, characterized in that the external additive is formed of an oxide fine particle including at least a silicon element, the oxide fine particle has a primary particle diameter R of 30 to 300 nm, a standard deviation δ of a particle size distribution of R is $R/4 \leq \delta \leq R$, and a shape of the external additive is substantially spherical with circularity SF1 and circularity SF2 of 100 to 130 and 100 to 15 125, respectively.

[0045]

(2) The external additive for electrophotographic toner according to (1), characterized in that the oxide fine particle includes at least the silicon element and a metal element.

20 [0046]

(3) The external additive for electrophotographic toner according to (1) or (2), characterized in that the oxide fine particle include at least the silicon element and a titanium element.

[0047]

25 (4) The external additive for electrophotographic toner according to (1), (2) or (3), characterized in that an element composition of the oxide fine particle is evenly dispersed between a surface part and an inside part of the oxide fine particle.

[0048]

30 (5) The external additive for electrophotographic toner according to (1), (2), (3), or (4), characterized in that the oxide fine particle is subjected to surface processing with at least an organosilicon compound.

[0049]

(6) The external additive for electrophotographic toner according to (1), (2), (3), (4), or (5), characterized in that the oxide fine particle is a hydrophobed oxide fine particle on which an $R^1_3SiO_{1/2}$ unit (R^1 is an identical or different monovalent hydrocarbon group each having 1 to 8 carbon atoms) is applied on a surface of the oxide fine particle.

[0050]

(7) The external additive for electrophotographic toner according to (1), (2), (3), (4), (5), or (6), characterized in that the oxide fine particle is subjected to surface processing by at least silicone oil and a liberation rate of the silicone oil is between 10 to 95 %.

[0051]

(8) Electrophotographic toner including at least a binder resin and a colorant and having a volume-average particle diameter of 2 to 7 μm , characterized in that the external additive for electrophotographic toner according to (1), (2), (3), (4), (5), (6), or (7) is mixed in the toner.

[0052]

(9) The electrophotographic toner, characterized in that the toner is mixed with the external additive for electrophotographic toner according to (1), (2), (3), (4), (5), (6), or (7) and one or more kinds of external additive of having a smaller average particle diameter of primary particles than the average particle diameter of primary particles of the external additive for electrophotographic toner.

[0053]

(10) The electrophotographic toner according to (8) or (9), characterized in that the binder resin of the toner includes at least a polyol resin.

[0054]

(11) The electrophotographic toner according to (8) or (9),

characterized in that the binder resin of the toner includes at least a polyester resin.

[0055]

(12) A two-component type developer, characterized by comprising
5 a carrier formed of at least the toner according to any one of (8) through (11) and a magnetic particle.

[0056]

(13) An image formation method, characterized by using the toner or the developer according to (8), (9), (10), (11), or (12).

10 [0057]

Description of the present invention is as follows. A publicly known external additive for toner, toner, and a production method and a material for a developer for use in the present invention, and a general system relating to an electrophotographic process
15 may be used in the present invention, provided that conditions are satisfied.

[0058]

(Oxide fine particle)

The oxide fine particle according to the present invention is a
20 spherical silica fine particle and may be produced by burning and decomposing alkoxysilane and/or partially hydrolyzed condensate of the alkoxysilane in a flame. The oxide fine particle is preferably a non-crystalline fine particle.

[0059]

25 The alkoxysilane is generally expressed by a formula $R^2_aSi(OR^3)_{4-a}$ (each of R^2 and R^3 is a monovalent hydrocarbon group having 1 to 4 carbon atoms, wherein "a" is an integral number of 0 to 4). Such alkoxysilanes include tetramethoxysilane, tetraethoxysilane, tetrapropoxysilane, tetrabutoxysilane, methyltrimethoxysilane, methyltriethoxysilane, methyltripropoxysilane, methyltributoxysilane,
30 ethyltrimethoxysilane, ethyltriethoxysilane, ethyltripropoxysilane, ethyltributoxysilane, propyltrimethoxysilane, propyltriethoxysilane, butyltrimethoxysilane, butyltriethoxysilane, dimethyldimethoxysilane,

dimethyldiethoxysilane, dimethyldipropoxysilane, dimethyldibutoxysilane, diethyldimethoxysilane, diethyldiethoxysilane, diethyldipropoxysilane, diethyldibutoxysilane, dipropyldimethoxysilane, dipropyldiethoxysilane, dibutyldimethoxysilane, dibutyldiethoxysilane, trimethylmethoxysilane, trimethylethoxysilane, trimethylpropoxysilane, trimethylbutoxysilane, triethylmethoxysilane, triethylethoxysilane, triethylpropoxysilane, triethylbutoxysilane, tripropylmethoxysilane, tripropylethoxysilane, tributylmethoxysilane, tributylethoxysilane, and so forth. Among them, the tetramethoxysilane and the methyltrimethoxysilane are preferred.

[0060]

The oxide fine particle is obtained by heating and evaporating an alkoxysilane and/or partially hydrolyzed condensate of the alkoxysilane and causing an evaporation product to flow into an inert gas such as a nitrogen gas, or by spraying the alkoxysilane and/or partially hydrolyzed condensate of the alkoxysilane into a flame such as an oxyhydrogen flame for burning and decomposition in the flame.

[0061]

To further improve environmental stability of charging of the oxide fine particle according to the present invention, the oxide fine particle is preferably a hydrophobed spherical silica fine particle of which an $R^1_3SiO_{1/2}$ unit is introduced on a surface of the oxide fine particle. R^1 is an identical or different monovalent hydrocarbon group each having 1 to 8 carbon atoms. Examples of such monovalent hydrocarbon groups are a methyl group, an ethyl group, a propyl group, a butyl group, a pentyl group, a hexyl group, a heptyl group, an octyl group, a cyclohexyl group, a phenyl group, a vinyl group, an allyl group, and so forth. Among them, the methyl group is particularly preferred.

[0062]

Introduction of $R^1_3SiO_{1/2}$ unit is performed according to a conventional method of surface modification of the silica fine

particle. That is, the $R^1_3SiO_{1/2}$ unit may be introduced by bringing a silazane compound generally expressed by a formula $R^1_3SiNHSiR^1_3$ into contact with a silica fine particle in the presence of water at 0 to 400°C in a gas, liquid, or solid phase, heating a resulting mixture at 50 to 400°C and thereby remove an excess of the silazane compound.

[0063]

Examples of the silazane compound generally expressed by the formula, $R^1_3SiNHSiR^1_3$, are hexamethyldisilazane, hexaethyldisilazane, hexapropyldisilazane, hexabutyldisilazane, hexapentyldisilazane, hexahexyldisilazane, hexacyclohexyldisilazane, hexaphenyldisilazane, divinyltetramethyldisilazane, and so forth. Among them, the hexamethyldisilazane is preferred for higher hydrophobicity of the modified silica fine particle and easier removal of the silazane compound.

[0064]

The electrostatic developer according to the present invention may be obtained by externally or internally adding the spherical silica fine particle to the toner particle. If the amount of spherical silica fine particle is less than 0.01 part by weight relative to 100 parts by weight of the toner, fluidity of the toner may be insufficient. If it is more than 20 parts by weight, a charging characteristic and a fixing characteristic of the toner are adversely affected. Therefore, the amount of the spherical silica fine particle is preferably in a range from 0.01 part by weight to 20 parts by weight, and more preferably from 0.1 part by weight to 5 parts by weight, relative to 100 parts by weight of the toner. The toner particle and the spherical silica fine particle may be mixed according to any given method using a device such as a V-type blender, a Henschel mixer, a ribbon blender, a Raikai mill, and so forth. The spherical silica fine particle may be adhered or fused to the surface of the toner particle, or be included in the toner particle.

[0065]

The oxide fine particle according to the present invention may preferably include a metal element (dopant compound), if needed, which is described below, as constitutional elements in addition to the silicon element (silicon compound). The compounds and oxides belonging to Groups II, III, and IV, and the third or higher period of the Periodic Table of Element are more preferred. Generally, elements such as Mg, Ca, Ba, Al, Ti, V, Sr, Zr, Si, Sn, Zn, Ga, Ge, Cr, Mn, Fe, Co, Ni, Cu, and so forth, are used. Among them, the Ti and the Zn are further preferred.

[0066]

The oxide fine particle according to the present invention is characterized by having a primary particle diameter R within a range of 30 to 300 nm, or more preferably within a range of 50 to 170 nm, wherein a standard deviation δ of a particle size distribution of R is described by a formula $R/4 \leq \delta \leq R$, or more preferably be described by a formula $R/3 \leq \delta \leq 2/3 \times R$, and being substantially spherical with circularity SF1 of 100 to 130 (or more preferably 100 to 125) and circularity SF2 of 100 to 125 (or more preferably 100 to 120). The primary particle diameter of the oxide fine particle is a number-average particle diameter.

[0067]

The particle diameter of the inorganic fine particle for use in the present invention may be determined by using a size distribution analyzer using a dynamic light-scattering particle such as DLS-700 available from Otsuka Electronics Co., Ltd. and Coulter N4 available from Coulter Electronics, Inc. However, it is difficult to dissociate secondary aggregation of the inorganic fine particle after hydrophobing processing. Therefore, it is preferable to directly obtain a particle diameter from photographs obtained by a scanning electron microscope (SEM) or a transmission electron microscope (TEM). When the SEM is used, an inherent form of the particle may be deteriorated due to deposited platinum, and so forth.

Therefore, it is preferable to determine the particle diameter by reducing a thickness of a deposited film, if any, to about 1 nm, or the particle diameter is determined, without vapor deposition, at a low acceleration voltage (several eV to 10 keV) using a high-resolution field emission scanning electron microscope FE-SEM (such as S-5200 available from Hitachi, Ltd.) which has sufficient resolution even when an acceleration voltage is lowered. When the SEM or the TEM is used, at least 100 oxide fine particles are to be observed, and the particle size distribution and the circularities SF1 and SF2 are to be statistically determined by calculation using an image processing apparatus, image processing software, and so forth, such as Luzex. Values determined by analysis performed by an image analyzing apparatus available from NIRECO Corporation (Luzex AP) and by calculation according to following equations are defined as SF-1 and SF-2. The values of SF-1 and SF-2 are preferably obtained by using Luzex. However, it is not limited to FE-SEM and the image processing apparatus, as long as similar analysis results are obtained.

[0068]

$$SF - 1 = (L^2/A) \times (\pi/4) \times 100$$

$$SF - 2 = (P^2/A) \times (1/4\pi) \times 100$$

In the equations,

an absolute maximum length of toner is expressed as L;

a projected area of toner is expressed as A; and

a maximum perimeter of toner is expressed as P.

If a particle is completely spherical, the value obtained by either equation is 100. As the value becomes larger than 100, the spherical particles becomes amorphous. In addition, SF-1 and SF-2 are formation coefficient, wherein SF-1 indicates a shape (oval, spherical, and so forth) of the entire toner, and SF-2 indicates a magnitude of depressions and protrusions on a surface of the toner particle.

[0069]

A composition of the oxide fine particle is preferably evenly dispersed between a surface part and an inside part of the oxide fine particles. Whether or not the composition is evenly dispersed may be determined by subjecting the surface (a 1/4 particle diameter surface) part and an inside part of the oxide fine particle to an elementary analysis mapping using a transmission electron microscope having a scanning function and an elementary analysis mapping function (HD-2000 available from Hitachi, Ltd., for instance). When a ratio of the surface part to the inside part is from 0.6 to 1.4, the composition may be determined as being evenly dispersed within the oxide fine particles.

When a Ti element is mixed in SiO_2 , values of A/C and B/C determined by measurement of the inside part of the particles (calculated value corresponds to an relative amount of Si and Ti on a basis of O), wherein A expresses a counted number of detected Si elements, B expresses a counted number of Ti elements, and C expresses a counted number of O elements. On the other hand, values of A'/C' and B'/C' are determined in a vicinity of the surface in the similar manner. The ratio is then determined by dividing A'/C' by A/C (or by dividing B'/C' by B/C).

In addition, elementary particles in the oxide fine particles preferably have a uniform size. The term, "uniform size," as used herein is defined as the ratio of a maximum diameter to a minimum diameter in a mass of elements is less than 10:1. The diameters of the elementary particles may be preferably easily determined by using, for instance, image analyzing software. It is preferable to analyze about 200 masses of elements.

[0070]

Examples of surface processing agents for the oxide fine particles are silane coupling agents such as dialkyldihalogenosilane, trialkylhalogenosilane, alkyltrihalogenosilane, hexaalkyldisilazane, and so forth, a silylating agent, a silane coupling agent having a fluoroalkyl group, an organic titanate coupling agent, an aluminum

coupling agent, silicone oil; silicone varnish, and so forth. An organosilicon compound coupling agent and a hydrophobing agent are more preferred.

[0071]

5 (External additive)

In addition to the oxide fine particles, inorganic fine particles and hydrophobed inorganic fine particles may be used in combination with the oxide fine particles as the external additive. The external additive preferably includes one or more kinds of hydrophobed inorganic fine particles having an average particle diameter of primary particles of 1 to 100 nm and more preferably 5 to 70 nm. The external additive more preferably includes one or more kinds of hydrophobed inorganic fine particles having an average particle diameter of primary particles of 20 nm or less and one or more kinds of inorganic fine particles having an average particle diameter of primary particles of 30 nm or more. These fine particles preferably have a specific surface area of 20 m²/g to 500 m²/g as determined according to the Brunauer-Emmett-Teller (BET) method.

[0072]

20 Any publicly known fine particles may be used as long as they satisfy requirements. For instance, silica fine particles, hydrophobic silica, fatty acid metallic salts (zinc stearate, aluminium stearate, and so forth), metallic oxides (titania, alumina, tin oxide, antimony oxide, and so forth), fluoropolymers, and so forth, may be included in the external additive.

[0073]

Examples of particularly preferred additives may include fine particles of hydrophobed silica, titania, titanium oxide, and aluminum. Such silica fine particles are available under trade names of HDK H 2000, HDK H 2000/4, HDK H 2050EP, HVK21, and HDK H 1303 (available from Hoechst AG), R972, R974, RX200, RY200, R202, R805, and R812 (available from Nippon Aerosil Co., Ltd.), and so forth. Titania fine particles are available under trade names of

P-25 (available from Nippon Aerosil Co., Ltd.), STT-30 and STT-65C-S (available from Titan Kogyo Kabushiki Kaisha), TAF-140 (available from FUJI TITANIUM INDUSTRY CO., LTD.), and MT-150W, MT-500B, MT-600B, MT-150A (available from TAYCA Corporation), and so forth.

5 Particularly, hydrophobed titanium oxide fine particles are available under trade names of T-805 (available from Nippon Aerosil Co., Ltd.), STT-30A, and STT-65S-S (available from Titan Kogyo Kabushiki Kaisha), TAF-500T and TAF-1500T (available from FUJI TITANIUM INDUSTRY CO., LTD.), MT-100S and MT-100T (available from
10 TAYCA Corporation), IT-S (available from Ishihara Sangyo Kaisha, Ltd.), and so forth.

[0074]

Such hydrophobed oxide fine particles, silica fine particles, titania fine particles, and alumina fine particles may be obtained
15 by treating hydrophilic fine particles with a silane coupling agent. Such silane coupling agents include methyltrimethoxysilane, methyltriethoxysilane, octyltrimethoxysilane, and so forth. In addition, silicone oil-processed oxide fine particles and the inorganic fine particles are also preferred. Such processed fine
20 particles are obtained by treating fine particles with silicon oil, if necessary, by heating.

[0075]

Such silicone oils include dimethyl silicone oil, methyl phenyl silicone oil, chlorophenyl silicone oil, methyl hydrogen silicone
25 oil, alkyl-modified silicone oil, fluorine-modified silicone oil, polyether-modified silicone oil, alcohol-modified silicone oil, amino-modified silicone oil, epoxy-modified silicone oil, epoxy-polyether-modified silicone oil, phenol-modified silicone oil, carboxyl-modified silicone oil, mercapto-modified silicone
30 oil, acrylic or methacrylic-modified silicone oil, alpha methylstyrene-modified silicone oil, and so forth. Examples of the inorganic fine particles are fine particles of silica, alumina, titanium oxide, barium titanate, magnesium titanate, calcium

titanate, strontium titanate, iron oxide, copper oxide, zinc oxide, tin oxide, silica sand, clay, mica, wollastonite, diatomaceous earth, chromium oxide, cerium oxide, iron oxide red, antimony trioxide, magnesium oxide, zirconium oxide, barium sulfate, barium carbonate, calcium carbonate, silicon carbide, silicon nitride, and so forth. Among them, silica and titanium dioxide fine particles are preferred. An amount to be added is preferably 0.1% by weight to 5% by weight, and more preferably 0.3% by weight to 3% by weight based on weight of the toner. The inorganic fine particles should preferably have an average particle diameter of primary particles of 100 nm or less, and more preferably 3 to 70 nm. If the average particle diameter is less than the diameter in range, the inorganic fine particles are buried in the toner and thereby fail to exhibit their functions effectively. If it is larger than the diameter in the range, the inorganic fine particles may unevenly damage a surface of photoconductor.

[0076]

The liberation rate of the silicone oil for use in the present invention may be determined by a following assay method. However, the determination method is not specifically limited to the assay method and any suitable method may be employed.

1. Extraction of free silicone oil

Dip a sample in chloroform, stir, and then leave the sample as it is. After removing supernatant liquid by centrifugation, remained solid matters are diluted with chloroform, the mixture is stirred and is then left to stand. This procedure is repeated to remove the free silicone oil.

2. Carbon content assay

The carbon content is determined by a CHN elementary analyzer (for instance, CHN Corder MT-5 [available from Yanaco Analytical Instruments Corporation]).

3. Determination of liberation rate of silicone oil

The liberation rate of the silicone oil is determined according

to a following equation.

[0077]

Silicone oil liberation rate = $(C_0 - C_1)/C_0 \times 100$ (%)

C_0 : the carbon content in the sample before extraction operation

5 C_1 : the carbon content in the sample after the extraction operation

(Surface treatment agent)

Examples of the surface treatment agents for the external additives including oxide fine particles are silane coupling agents such as dialkyldihalogenosilane, trialkylhalogenosilane, alkyltrihalogenosilane, 10 hexaalkyldisilazane, and so forth, silylating agents, silane coupling agents having a fluoroalkyl group, organotitanate coupling agents, aluminum coupling agents, silicone oil, silicone varnish, and so forth. Organosilicon compound surface treatment agents and hydrophobing processing agents are more preferred.

15 [0078]

(Softening point, flow beginning temperature)

The softening point and the flow beginning temperature of the toner according to the present invention is determined by using a softening point measuring apparatus (FP90 available from Mettler 20 Toledo GmbH) at a heating rate of 1 °C / min.

[0079]

(Glass transition temperature (T_g))

The glass transition temperature (T_g) of the toner according to the present invention is determined by using a following 25 differential scanning calorimeter under following conditions.

[0080]

- Differential scanning calorimeter: SEIKO1DSC100

SEIKO1SSC5040 (Disk Station)

- Determination conditions:

30 Temperature range: 25 to 150 °C

Heating rate: 10 °C/min

Sampling time: 0.5 sec

Sample amount: 10 mg

(Molecular weight)

Determination of the number-average molecular weight (M_n), weight-average molecular weight (M_w), and peak molecular weight (M_p) are performed by GPC (gel permeation chromatography) in a following manner. 80 mg of a sample is dissolved in 10 ml of THF to yield a sample solution, and the sample solution is filtrated through a 5 μ m-filter. 100 μ l of the sample solution is then injected into a column, and retention time of the sample is determined under following conditions. In addition, the retention time is determined by using polystyrene having a known average molecular weight as a reference material. Then a number-average molecular weight of the sample in terms of polystyrene is determined based on a calibration curve prepared before hand.

[0081]

- Column: Guard column + GLR400M + GLR400M + GLR 400
(all available from Hitachi, Ltd.)
- Column temperature: 40 °C
- Mobile phase (flow rate): THF (1 ml/min)
- Peak detection method: UV (254 nm)

[0082]

(Needle penetration degree, thermal storage stability)

Toner weighed per each 10 g is placed in a 20 cc glass container and is left to stand in a thermostatic chamber set at 50 °C for 5 hours. Then the needle penetration degree of a sample is determined by a penetrometer.

[0083]

(Average particle diameter of dispersed wax)

The average particle diameter of dispersed wax relating to the present invention may be analyzed by observing ultra-thin sections of the toner by using a TEM (transmission electron microscope). If necessary, the observed TEM images are input in a computer, and the average particle diameter of the dispersed wax is determined by image processing software. Instead of TEM, any other means such

as an optical microscope, a CCD camera, and a laser microscope may be used, as long as they may determine the average particle diameter.

[0084]

(Binder resin)

5 Binder resins according to the present invention include polymers of styrene and polymers of substituted styrenes such as polystyrenes, poly-p-chlorostyrenes, polyvinyltoluenes, and so forth; styrenic copolymers such as styrene-p-chlorostyrene copolymers, styrene-propylene copolymers, styrene-vinyltoluene
10 copolymers, styrene-vinylnaphthalene copolymers, styrene-methyl acrylate copolymers, styrene-ethyl acrylate copolymers, styrene-butyl acrylate copolymers, styrene-octyl acrylate copolymers, styrene-methyl methacrylate copolymers, styrene-ethyl methacrylate copolymers, styrene-butyl methacrylate copolymers,
15 styrene-methyl alpha-chloromethacrylate copolymers, styrene-acrylonitrile copolymers, styrene-vinyl methyl ketone copolymers, styrene-butadiene copolymers, styrene-isoprene copolymers, styrene-acrylonitrile-indene copolymers, styrene-maleic acid copolymers, styrene-maleic ester copolymers, and so forth;
20 polymethyl methacrylate, polybutyl methacrylate, poly vinyl chloride, poly vinyl acetate, polyethylene, polypropylene, polyester, an epoxy resin, a polyol resin, polyurethane, polyamide, poly vinyl butyral, a poly acrylic acid resin, rosin, modified rosin, a terpene resin, an aliphatic or an alicyclic hydrocarbon resin,
25 an aromatic petroleum resin, chlorinated paraffins, paraffin wax, and so forth. Each of these resins may be used either alone or in combination. Among them, particularly the polyester resin and the polyol resin are more preferred.

[0085]

30 The binder resin more preferably includes a polyol resin or a polyol resin having an epoxy resin moiety and a polyoxyalkylene moiety in its main chain. Thus, a sufficient compressive strength, a tensile rupture strength, environmental stability, and a stable

fixing characteristic are achieved. Further, a toner image may be prevented from transferring onto a vinyl chloride resin sheet when a surface of a fixed image of a copy is closely contacting the sheet. Particularly, when the above-mentioned resins are used in color toner, it is favorably effective for color reproducibility, stable glossiness, and in preventing the fixed image of copy from curling. Further, by including at least a polyol resin moiety and a polyester resin moiety, the resulting toner has a well-balanced stretching characteristic and an adhesion characteristic as well as an improved compressive strength, and a further stable transfer characteristic, a development characteristic and an image-fixing characteristic may be favorably achieved.

[0086]

Epoxy resins for use in the present invention are preferably epoxy resins obtained by combining bisphenol such as bisphenol A and bisphenol F with epichlorohydrin. More preferably, the epoxy resin includes two or more kinds of bisphenol A epoxy resin having different number-average molecular weights to achieve a stable fixing characteristic and glossiness. A low molecular weight component of the epoxy resin preferably has a number-average molecular weight of 360 to 2000, and a high molecular weight component of the epoxy resin preferably has a number-average molecular weight of 3000 to 10000. Further, the low molecular weight component of the epoxy resin is preferably 20 to 50 wt % and the high molecular weight component of the epoxy resin is preferably 5 to 40 wt %. If the epoxy resin includes an excessively large amount of the low molecular weight component or includes a low molecular weight component having a low number-average molecular weight of less than 360, the resulting toner may have excessive glossiness or deteriorated storage stability. If the epoxy resin includes an excessively large amount of the high molecular weight component or includes a high molecular weight component having a high number-average molecular weight more than

N-methyl(ethyl)piperazine, piperidine, and so forth. The carboxylic acids include a propionic acid, a caproic acid, and so forth.

[0090]

5 The polyol resin having an epoxy resin moiety and an alkylene oxide moiety in its main chain according to the present invention may be obtained from various combinations of materials. For instance, the polyol resin maybe obtained by causing an epoxy resin having glycidyl groups at both ends and an alkylene oxide adduct
10 of a dihydric phenol having glycidyl groups at both ends to react with dihalide, diisocyanate, diamine, dithiol, polyhydric phenol, or a dicarboxylic acid. Among them, dihydric phenol is preferably used for a more stable reaction. It is also preferable to use polyhydric phenol and a polycarboxylic acid in combination with
15 the dihydric phenol within ranges not inviting gelation. An amount of the polyhydric phenol and the polycarboxylic acid is preferably 15% by weight or less, and more preferably 10% by weight or less, relative to the total amount of the materials.

[0091]

20 Examples of the compounds having two or more active hydrogens in the molecule according to the present invention include dihydric phenols, polyhydric phenols, and a polycarboxylic acid. Such dihydric phenols include bisphenol such as bisphenol A and bisphenol F. Such polyhydric phenols include o-cresol novolacs, phenol
25 novolacs, tris (4 - hydroxyphenyl)methane, 1 - [alpha - methyl-alpha - (4 - hydroxyphenyl) ethyl]benzene, and so forth. Examples of the polycarboxylic acids are a malonic acid, a succinic acid, a glutaric acid, an adipic acid, a maleic acid, a fumaric acid, a phthalic acid, a terephthalic acid, a trimellitic acid, a
30 trimellitic anhydride, and so forth.

[0092]

The resin according to the present invention more preferably includes a polyol resin having an epoxy resin moiety, a

polyoxyalkylene moiety, and a polyester moiety in a main chain in the resin. Thus, viscoelasticity and rigidity of the resin are changed particularly due to a polyester component. Accordingly, a more flexible physical property of the resin may be achieved and image curling may be favorably prevented from occurring.

[0093]

The binder resins preferably have an amount of controlled epoxy equivalent of 10000 or more, more preferably 30000 or more, and still more preferably 50000 or more. Thus, a heat characteristic of the resin may be controlled and an amount of low molecular epichlorohydrin, or a reaction residue, and so forth, may be reduced. Accordingly, toner with excellent safeness and a resin characteristic may be achieved.

[0094]

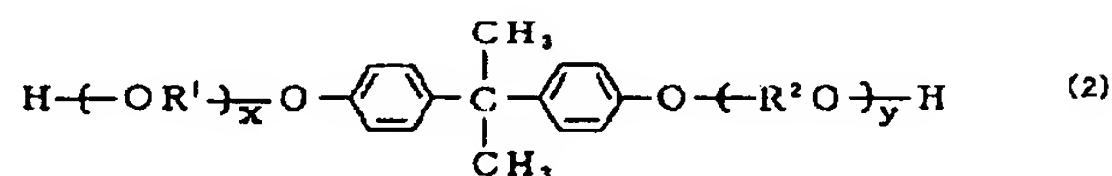
Various types of polyester resins may be used in the present invention. However, the polyester resins are preferably obtained by reaction among following components ①, ②, and ③.

① at least one type of polyester resin selected from dicarboxylic acid, lower alkyl ester of the dicarboxylic acid, and acid anhydride;

② a diol component expressed by a following formula (2):

[0095]

[Chemical formula 2]



(in the formula, R^1 and R^2 may be identical or different and each expresses an alkylene group containing 2 to 4 carbon atoms; x and y each expresses a number of a repeated unit of 1 or more; and $x + y = 2$ to 16)

[0096]

③ at least one type of polyester resin selected from trivalent or higher polycarboxylic acids, lower alkyl esters and acid anhydrides of the trivalent or higher polycarboxylic acids, and

trihydric or higher polyhydric alcohols.

[0097]

Examples of the component ①, that is, a dicarboxylic acid, lower alkyl ester of the dicarboxylic acid, and acid anhydrides of the dicarboxylic acid, are a terephthalic acid, an isophthalic acid, 5 a sebacic acid, an isodecylsuccinic acid, a maleic acid, and a fumaric acid, monomethyl, monoethyl, dimethyl, and diethyl esters of these carboxylic acids, phthalic anhydride, maleic anhydride, and so forth. Among them, the terephthalic acid, the isophthalic 10 acid, and the dimethyl esters thereof are preferred for higher blocking resistance and lower cost. These dicarboxylic acids, lower alkyl esters and acid anhydrides thereof largely affect the fixing characteristic and blocking resistance of the toner. That is, although it depends on a condensation degree, when a large amount 15 of an aromatic carboxylic acid such as a terephthalic acid or an isophthalic acid is used, it decreases the fixing characteristic although it increases the blocking resistance. In contrast, when a large amount of a sebacic acid, an isodecylsuccinic acid, a maleic acid, a fumaric acid, and so forth, is used, the blocking resistance 20 is decreased, although it increases the fixing characteristic. Accordingly, these dicarboxylic acids are appropriately selected and used alone or in combination according to the composition of the other monomers, proportions, and a degree of condensation.

[0098]

25 Examples of the diol component ② expressed by the formula (I) are polyoxypropylene - (n) - polyoxyethylene - (n') - 2, 2 - bis (4 - hydroxyphenyl) propane, polyoxypropylene - (n) - 2, 2 - bis (4 - hydroxyphenyl) propane, polyoxyethylene - (n) - 2, 2 - bis (4 - hydroxyphenyl) propane, and so forth. Among them, 30 polyoxypropylene - (n) - 2, 2 - bis (4 - hydroxyphenyl) propane where $2.1 \leq n \leq 2.5$, and polyoxyethylene - (n) - 2, 2 - bis (4 - hydroxyphenyl) propane where $2.0 \leq n \leq 2.5$ are preferred. These diol components are advantageous to increase a glass transition

temperature and to control the reaction more easily.

[0099]

As the diol component, aliphatic diols such as ethylene glycol, diethylene glycol, 1, 2 - butanediol, 1, 3 - butanediol, 1, 4 -
5 butanediol, neopentyl glycol, propylene glycol, and so forth, may also be used.

[0100]

Examples of the trivalent or higher polycarboxylic acids, lower alkyl esters and acid anhydrides of the trivalent or higher
10 polycarboxylic acids according to ③ include 1, 2, 4 - benzenetricarboxylic acid (trimellitic acid), 1, 3, 5 - benzenetricarboxylic acid, 1, 2, 4 - cyclohexanetricarboxylic acid, 2, 5, 7 - naphthalenetricarboxylic acid, 1, 2, 4 - naphthalene -
15 - hexanetricarboxylic acid, 1, 3 - dicarboxy - 2 - methyl - 2 - methylenecarboxypropane, tetra (methylenecarboxy) methane, 1, 2, 7, 8 - octanetetracarboxylic acid, empol trimer acid, and monomethyl, monoethyl, dimethyl, diethyl esters of these polycarboxylic acids, and so forth.

20 [0101]

Examples of the trihydric or higher polyhydric according to ③ are sorbitol, 1, 2, 3, 6 - hexanetetrol, 1, 4 - sorbitan, pentaerythritol, dipentaerythritol, tripentaerythritol, sucrose, 1, 2, 4 - butanetriol, 1, 2, 5 - pentanetriol, glycerol, diglycerol,
25 2 - methylpropanetriol, 2 - methyl - 1, 2, 4 - butanetriol, trimethylolethane, trimethylolpropane, 1, 3, 5 - trihydroxy methylbenzene, and so forth.

[0102]

An amount of the trivalent or higher polyvalent monomers is
30 preferably 1% by mole to 30% by mole relative to the total amount of the monomer composition. If the amount is 1% by mole or less, the toner may have a decreased anti-offset characteristic and deteriorated durability. If it is 30% by mole or more, the toner

may have a deteriorated fixing characteristic.

[0103]

Among these trivalent or higher polyvalent monomers, benzenetricarboxylic acids, and anhydrides, esters, and other derivatives of the benzenetricarboxylic acids are particularly preferred. That is, by using the benzenetricarboxylic acids, both of the fixing characteristic and the anti-offset characteristic may be satisfied at the same time.

[0104]

10 These polyester resins and polyol resins are preferably not crosslinked or are weakly crosslinked (preferably have a content of THF-insoluble matters of 5% or less). If they are highly crosslinked, it is difficult to achieve satisfactory transparency and glossiness. A production method of the binder resin is not particularly limited, but the binder resin may be produced according to any procedure such as bulk polymerization, solution polymerization, emulsion polymerization, suspension polymerization, and so forth.

[0105]

20 (Colorant)

Any publicly known dyes and pigments may be used as the colorant for the toner according to the present invention. Such dyes and pigments include, but are not limited to, carbon black, nigrosine dyes, black iron oxide, Naphthol Yellow S, Hansa Yellow (10G, 5G, and G), cadmium yellow, yellow iron oxide, yellow ochre, chrome yellow, Titan Yellow, Oil Yellow, Hansa Yellow (GR, A, RN, and R), Pigment Yellow L, Benzidine Yellow (G, GR), Permanent Yellow (NCG), Vulcan Fast Yellow (5G, R), Tartrazine Lake, Quinoline Yellow Lake, Anthragen Yellow BGL, isoindolinone yellow, red oxide, red lead oxide, red lead, cadmium red, cadmium mercury red, antimony red, Permanent Red 4R, Para Red, Fire Red, p-chloro-o-nitroaniline red, Lithol Fast Scarlet G, Brilliant Fast Scarlet, Brilliant Carmine BS, Permanent Red (F2R, F4R, FRL, FRLL, F4RH), Fast Scarlet VD,

Vulcan Fast Rubine B, Brilliant Scarlet G, Lithol Rubine GX, Permanent Red F5R, Brilliant Carmine 6B, Pigment Scarlet 3B, Bordeaux 5B, Toluidine Maroon, Permanent Bordeaux F2K, Helio Bordeaux BL, Bordeaux 10B, BON Maroon Light, BON Maroon Medium, eosine lake, Rhodamine Lake B, Rhodamine Lake Y, Alizarine Lake, Thioindigo Red B, Thioindigo Maroon, Oil Red, quinacridone red, Pyrazolone Red, Chrome Vermilion, Benzidine Orange, Perynone Orange, Oil Orange, cobalt blue, cerulean blue, Alkali Blue Lake, Peacock Blue Lake, Victoria Blue Lake, metal-free phthalocyanine blue, Phthalocyanine Blue, Fast Sky Blue, Indanthrene Blue (RS, BC) , indigo, ultramarine, Prussian blue, Anthraquinone Blue, Fast Violet B, Methyl Violet Lake, cobalt violet, manganese violet, dioxazine violet, Anthraquinone Violet, chrome green, zinc green, chromium oxide, viridian emerald green, Pigment Green B, Naphthol Green B, Green Gold, Acid Green Lake, Malachite Green Lake, Phthalocyanine Green, Anthraquinone Green, titanium oxide, zinc white, lithopone, mixtures of the above-mentioned dyes, and so forth. An amount of the colorant is generally from about 0.1 part by weight to 50 parts by weight relative to 100 parts by weight of the binder resin.

[0106]

(Master batch pigment)

A colorant for use in the present invention may be a master batch pigment prepared in advance by mixing and kneading a pigment with a resin in nearly equal proportions in order to improve affinity of the resin and the pigment. More preferably, a master batch pigment having excellent environmentally-related charging stability may be prepared by heating and kneading the pigment and a resin containing components soluble in a low polar solvent without use of an organic solvent. Dispersibility of the master batch pigment may be further improved by using a dry powder pigment and using water as a way of moistening the dry powder pigment with the resin. An organic pigment used as a colorant is generally hydrophobic, but water may be penetrated into an inside part of

an aggregation of the pigments by applying some force, since the organic pigment is subjected to a water washing process and a drying processes during its manufacture. When the pigment containing water inside the aggregation is kneaded with a resin in an open kneader at temperature of 100 °C or higher, water inside the aggregation reaches its boiling point at once and expands, causing force to crush the aggregation from inside the aggregation. The force from inside the aggregation may very efficiently crush the aggregation compared to a force applied externally.

[0107]

In this case, the resin is heated to a temperature higher than its softening point. Therefore, viscosity is decreased and the aggregation may be efficiently moistened. In addition, the resin is replaced with the water heated at temperature around its boiling point inside the aggregate due to an effect similar to so-called flashing. Accordingly, the master batch pigments containing pigments dispersed in a state close to primary particles. In a process of water vaporization, the water deprives the kneaded product of the heat of vaporization, and the kneaded product is held at a relatively low temperature of 100 °C or lower at relatively high viscosity. Thus, a shearing force is effectively applied to the aggregation of the pigment. Open kneaders for use in manufacture of the master batch pigments include regular two-roll kneaders, and three-roll kneaders, as well as open-type Banbury mixers. Further, continuous two-roll kneaders available from Mitsui Mining Co., Ltd., and so forth, may be used.

[0108]

(Charge control agent)

The toner according to the present invention may further include a charge control agent if necessary. Any publicly known charge control agents may be used, for instance, nigrosine dyes, triphenylmethane dyes, chromium-containing metal complex dyes, molybdic acid chelate pigments, rhodamine dyes, alkoxyamines,

quaternary ammonium salts (including fluorine-modified quaternary ammonium salts), alkylamides, simple substance or compounds of phosphorus, simple substance or compounds of tungsten, fluorine-containing active agents, metal salts of salicylic acid, metal salts of salicylic acid derivatives, and so forth. Specific examples of the charge control agents include BONTRON 03 of Nigrosine dyes, BONTRON P-51 of quaternary ammonium salt, BONTRON S-34 of metal-containing azo dye, BONTRON E-82 of metal complex of oxynaphthoic acid, BONTRON E-84 of metal complex of salicylic acid, and BONTRON E-89 of phenolic condensation product (available from Orient Chemical Industries Co., Ltd.), TP-302 and TP-415 of molybdenum complex of quaternary ammonium salt (available from Hodogaya Chemical Co., Ltd.), COPY CHARGE PSY VP2038 of quaternary ammonium salt, COPY BLUE PR of triphenylmethane derivative, COPY CHARGE NEG VP2036 and COPY CHARGE NX VP434 of quaternary ammonium salt (available from Hoechst AG), LRA-901 and LR-147 of boron complex (available from Japan Carlit Co., Ltd.), as well as copper phthalocyanine pigments, perylene pigments, quinacridone pigments, azo pigments, and polymeric compounds having a functional group such as a sulfonic group, a carboxyl group, quaternary ammonium salt, and so forth.

[0109]

In the present invention, an amount of the charge control agent is not specifically limited to, but is determined according to a type of a binder resin, an additive, if any, used according to need, and a production method of the toner including a dispersion method. However, the amount of the charge control agent is preferably from 0.1 part by weight to 10 parts by weight, and more preferably from 2 parts by weight to 5 parts by weight, relative to 100 parts by weight of the binder resin. If the amount is more than 10 parts by weight, the toner may have excessively high charges and an effect of a main charge control agent may be reduced. Accordingly, electrostatic attraction to a development roller is increased,

causing decreased fluidity of the developer or decreased image density.

[0110]

(Carrier)

5 The toner according to the present invention may be used in a two-component developer by mixing with a magnetic carrier. An amount of the toner in the developer is preferably from 1 part by weight to 10 parts by weight relative to 100 parts by weight of the carrier. As to such magnetic carriers, conventional magnetic
10 particles with a particle diameter of about 20 μm to 200 μm , made of iron powder, ferrite powder, magnetite powder, a magnetic resin carrier, and so forth, may be used. Such coating materials include an amine resin such as an urea-formaldehyde resin, a melamine resin, a benzoguanamine resin, a urea resin, a polyamide resin, an epoxy
15 resin, and so forth. Further, a halogenated olefin resin such as a polyvinyl resin and a polyvinylidene resin including an acrylic resin, a polymethyl methacrylate resin, a polyacrylonitrile resin, a polyvinyl acetate resin, a polyvinyl alcohol resin, a polyvinyl butyral resin, a polystyrene resin, a styrene-acrylic copolymer
20 resin, polyvinyl chloride, and so forth, a polyester resin such as a polyethylene terephthalate resin, a polybutylene terephthalate resin, a polycarbonate resin, a polyethylene resin, a polyvinyl fluoride resin, a polyvinylidene fluoride resin, a polytrifluoroethylene resin, a polyhexafluoropropylene resin,
25 copolymers of vinylidene fluoride and acrylic monomer, vinylidene fluoride-vinyl fluoride copolymers, terpolymers of tetrafluoroethylene, vinylidene fluoride, a non-fluorinated monomer, other fluoroterpolymers, and a silicone resin may be used.

[0111]

30 Thickness of a film formed of the above-mentioned coating materials is preferably 0.01 μm to 3 μm , and more preferably 0.1 μm to 0.3 μm . If the thickness is 0.01 μm or less, it is difficult to control the film and a function as a coating film may not be

performed. Further, if the thickness is more than 3 μm , no conductivity may be unfavorably obtained. The resin for use in the coating material may further include a conductive powder, and so forth, according to need. Such conductive powders include a metal powder, carbon black, titanium oxide, tin oxide, zinc oxide, and so forth. These conductive powders preferably have an average particle diameter of 1 μm or less. If the average particle diameter is more than 1 μm , it is difficult to control electric resistance.

[0112]

10 The toner according to the present invention may be used as one-component magnetic toner in which a carrier is not used or as nonmagnetic toner.

[0113]

(Magnetic material)

15 Further, the toner according to the present invention may be used as magnetic toner by including the magnetic material. To use the toner as the magnetic toner, fine particles of a magnetic substance may be included in the toner. Such magnetic substances include ferromagnetic metals such as iron including ferrite and magnetite, cobalt, nickel, and so forth, and alloys of the magnetic substances, compounds containing elements of the magnetic substances, alloys which do not contain a ferromagnetic element but show ferromagnetism by being subjected to an appropriate heat processing, such as whistler alloys containing manganese and copper such as manganese-copper-aluminum alloys and manganese-copper-tin alloys, chromium dioxide, and so forth. The magnetic substance is preferably uniformly dispersed in the toner in a form of a fine powder having an average particle diameter of 0.1 μm to 1 μm . An amount of the magnetic material is preferably from 10 parts by weight to 70 parts by weight, and more preferably from 20 parts by weight to 50 parts by weight, relative to 100 parts by weight of the toner.

[0114]

(Wax)

The toner or the developer preferably includes wax to have a fixing-releasing characteristic. Particularly, when an oilless fixing device in which oil application on an image fixing part is not performed is used, it is preferable to include the wax in the toner. The wax has a melting point of preferably from 40 to 120 °C and more preferably from 50 to 110 °C. If the wax has an excessively high melting point, the toner may have an insufficient fixing characteristic at a low temperature. On the other hand, if the wax has an excessively low melting point, the toner may have a decreased anti-offset characteristic and durability. The melting point of the wax may be determined by differential scanning calorimetry (DSC). More specifically, several mg of a sample is heated at a constant heating rate, such as 10 °C/min, and the melting peak obtained in the procedure is defined as the melting point. A content of the wax is preferably from 0 part by weight to 20 parts by weight, and more preferably from 0 part by weight to 10 parts by weight.

[0115]

Wax which may be used in the present invention may include solid paraffin wax, microcrystalline wax, rice wax, fatty acid amide wax, fatty acid wax, aliphatic monoketones, fatty acid metal salt wax, fatty acid ester wax, partially saponified fatty acid ester wax, silicone varnish, higher alcohol, carnauba wax, and so forth. In addition, polyolefins such as low molecular weight polyethylenes, polypropylenes may be used as the wax. Among them, polyolefins and esters having a softening point of 60 to 150 °C, and more preferably 70 to 120 °C according to a ball and ring method are preferred.

[0116]

It is proved to be effective that the toner more preferably includes at least one wax selected from free-fatty-acid-free type carnauba wax having an acid value of 5 mgKOH/g or less, montan ester wax, oxidized rice wax having an acid value of 10 mgKOH/g to 30 mgKOH/g, and sasol wax. Such free-fatty-acid-free type carnauba wax is prepared by removing a free fatty acid from material carnauba

wax and have an acid value of 5 mgKOH/g or less, accordingly, and is micro crystallite with smaller particles than conventional carnauba wax and may be dispersed in a binder resin in a form of particle having an average particle diameter of 1 μ m or less.

5 Accordingly, dispersibility of the free-fatty-acid-free type carnauba wax is improved. The montan ester wax is purified from minerals, have a smaller particle diameter, and may be dispersed in the binder resin in a form of particle having an average particle diameter of 1 μ m or less as in the carnauba wax. Particularly,
10 the montan ester wax preferably has an acid value of 5 mgKOH/g to 14 mgKOH/g.

[0117]

The dispersed particles of the wax have a diameter of preferably 3 μ m or less, more preferably 2 μ m or less, and still more
15 preferably 1 μ m or less. When the dispersed particles have a diameter more than 3 μ m, the resulting toner may have deteriorated durability at a high temperature and high humidity and a decreased charging stability, although wax flowability and releasability of a transferring material are improved.

20 [0118]

Oxidized rice wax is prepared by oxidizing rice bran wax with the air. The oxidized rice bran wax preferably has an acid value of 10 mgKOH/g to 30 mgKOH/g. If the acid value is less than 10 mgKOH/g, a lower limit temperature for image-fixing may rise.
25 Accordingly, a fixing characteristic at a low temperature may not be sufficiently achieved. If it is more than 30 mgKOH/g, a cold-offset temperature increases. Accordingly, the fixing characteristic at a low temperature may not be sufficiently achieved. Examples of the sasol wax are Sasol Wax H1, H2, A1, A2, A3, A4,
30 A6, A7, A14, C1, C2, SPRAY30, SPRAY40, and so forth, available from Sasol Co., Ltd. Among them, Sasol Wax H1, H2, SPRAY30, and SPRAY 40 are preferred for their good image-fixing properties at a low temperature and good storage stability. Each of the wax may be used

either alone or in combination. By using the wax in an amount of from 1 part by weight to 15 parts by weight, and more preferably from 2 parts by weight to 10 parts by weight, relative to 100 parts by weight of the binder resin, an above-mentioned favorable result
5 may be obtained.

[0119]

(Cleaning improver)

It is further preferable to include the cleaning improver for removing the developer remained on a photoconductor or a primary
10 transfer medium after transfer in the toner or the developer, or to apply the cleaning improver on a surface of a toner particle or a developer particle. The cleaning improver may include fatty acidmetallic salts such as zinc stearate, calcium stearate, stearic acid, and fine polymer particles prepared by, for instance,
15 soap-free emulsion polymerization such as polymethyl methacrylate fine particles and polystyrene fine particles. Such fine polymer particles preferably have a relatively narrow particle size distribution and a volume-average particle diameter of 0.01 μm to 1 μm . The amount of the cleaning improver is preferably from 0.001
20 part by weight to 5 parts by weight and more preferably from 0.001 part by weight to 1 part by weight.

[0120]

(Production method of toner)

The production method of toner including a mixing process of
25 mechanically mixing a developer component including at least a binder resin, a main charge control agent, and a pigment, a melting and kneading process, a pulverizing process, and a classifying process, may be used as the production method of toner according to the present invention. Further, the production method of toner
30 may further include a process in which powders other than product particles obtained in the pulverizing process and the classifying process are returned and used in the mixing process or the melting and kneading process.

[0121]

The term other powders than the product particles (by-products) as described herein means fine particles or crude particles other than the product component having a desired particle diameter obtained in the pulverizing process after the melting and kneading process, or fine particles or crude particles other than the product component having a desired particle diameter generated in the subsequent classifying process. These by-products are preferably mixed with the raw materials in a mixing process or in the melting and kneading process. The weight ratio of the by-products to the raw materials is preferably 1:99 to 50:50.

[0122]

The mixing process in which a component of the developer including at least the binder resin, the main charge control agent, the pigment and the by-products is mechanically mixed, is not limited and may be performed by using a regular mixing apparatus such as an apparatus with a rotating blade under usual conditions.

[0123]

When the mixing process is completed, the resulting mixture is then charged into a kneader and is subjected to melting and kneading. Such melting kneaders include a single-screw or a twin-screw continuous kneader, a roll-mill batch-system kneader, and so forth. For instance, a twin-screw extruding machine Model KTK from Kobe Steel Co., Ltd., a TEM series extruding machine from TOSHIBA MACHINE Co., Ltd., a twin-screw extruding machine from KCK Tool & Die, Co., a twin-screw extruding machine Model PCM from Ikegai, Ltd., and a co-kneader from Buss Co., Ltd. may be favorably used.

[0124]

The melting and kneading process must be performed under appropriate conditions so as not to cause cleavage of molecular chains of the binder resin. More specifically, a temperature for melting and kneading is to be set based on a softening point of the binder resin. If it is excessively lower than the softening

point, the molecular chains of the binder resin are significantly cleaved. If it is excessively higher than the softening point, sufficient dispersion may not be preformed. To control an amount of volatile components in the toner, it is preferable to set optimum
5 conditions on the temperature, time, and atmosphere during melting and kneading while monitoring an amount of residual volatile components.

[0125]

After completing the melting and kneading process, the resulting
10 kneaded product is pulverized. In the pulverizing process, it is preferable to roughly pulverize the kneaded product and then finely pulverize the roughly pulverized product. In this case, a method of pulverizing in which the product is caused to collide with a collision board in a jet stream or a method of pulverizing in which
15 the product is pulverized in a narrow gap between a mechanically rotating rotor and a stator is preferably used.

[0126]

After completing the pulverizing process, the pulverized product is classified in an airflow by a centrifugal force to produce toner
20 particles (base particle) having a predetermined particle diameter such as a volume-average particle diameter of 2 μm to 7 μm . The volume-average particle diameter may be determined by using, for instance, a COULTER TA-II (available from Coulter Electronics, Inc.) and so forth.

25 [0127]

In addition, when preparing the toner, the inorganic fine particles according to the present invention, such as the above-mentioned oxide fine particles and the hydrophobic silica fine particles may be added to the prepared toner to improve fluidity,
30 storage stability, a development characteristic, and a transfer characteristic of the toner. Regular mixing apparatus for powders is used for mixing the external additive. However, it is preferable that the mixing apparatus is having a jacket, and so forth, and

is capable of adjusting an inside temperature. To change a record of a load applied to the external additive, the external additive may be added in between the process or gradually. Alternatively, a number of rotations, a speed of rolling, time, and a temperature of the mixing apparatus may be changed to change the record of the load. It is acceptable that a relatively high load is applied at an early stage, and a relatively low load is then applied, or they may be applied in a reverse order.

[0128]

10 Examples of mixing systems for mixing equipment are a V mixer, a rocking mixer, a Ledige mixer, a nauta mixer, a Henshel mixer, and so forth. Another method such as a polymerization method or capsulation method may be used as a production method of toner. These methods are schematically described below.

15 [0129]

(Polymerization method 1)

- ① A polymerizable monomer, and where necessary, a polymerization initiator, a colorant, wax, and so forth, are granulated in a water-based disperse medium.
- 20 ② The granulated monomer composition particles are classified into an appropriate particle diameter.
- ③ The monomer composition particles having a specific particle diameter obtained by the classification are polymerized.
- ④ The dispersing agent is removed by appropriate processing, and
25 the resulting polymerization product is subjected to filtration, water washing, and drying to yield base toner particles.

[0130]

(Polymerization method 2)

- ① A low molecular weight resin, a high molecular weight resin,
30 a colorant, wax, a wax-dispersing agent, and where necessary a charge control agent, and so forth, are dispersed in an oily disperse medium using a solvent such as ethyl acetate, and so forth.
- ② The resulting dispersion liquid is then dropped in water

containing organic fine particles and an elongation agent for emulsification and convergence.

③ The resulting dispersion liquid is heated for polymerization and solvent removal.

5 ④ The resulting mixture is aged in water, and then washed, collected, dried to obtain base toner particles.

[0131]

(Capsulation method)

10 ① A resin, and where necessary a colorant, are kneaded by using a kneading device to obtain a molten toner core material.

② The toner core material is placed in water and is strongly agitated to form a core material in a form of a fine particle.

15 ③ The fine particles for the core material are placed in a solution of a shell material and agitated, and a poor solvent is added in the solution to cover a surface of the core material with the shell material for encapsulation.

④ The resulting capsules are filtrated and dried to obtain base toner particles.

[0132]

20 (Intermediate transfer body)

An intermediate transfer body may be used in the present invention. Embodiment 1 of an intermediate transfer body in a transfer system is described below. Fig. 1 is a schematic view illustrating a configuration of a copier relating to an embodiment of toner and a developer according to the present invention. Arranged around a photoconductive drum (hereinafter referred to as a photoconductor) 10 functioning as an image carrier are a charging roller 20 functioning as a charging device, an exposure device 30, a cleaning device 60 having a cleaning blade, a discharging lamp 90 functioning as a discharging device, development apparatus 40, and an intermediate transfer body 50 functioning as the intermediate transfer body. The intermediate transfer body 50 is suspended by a plurality of suspension rollers 51 and is designed to be endlessly

25
30

driven by a drive means such as a motor, not shown, in a direction of an arrow. One of the suspension rollers 51 serves as a transfer bias roller for applying transfer bias on the intermediate transfer body, wherein a predetermined transfer bias voltage is applied from a power source, not shown. In addition, a cleaning device 90 having a cleaning blade for the intermediate transfer body 50 is arranged. A transfer roller 80 functioning as a transfer means for transferring an image to be developed on transfer paper 100 functioning as a final transfer material is arranged facing the intermediate transfer body 50. The transfer bias is applied to the transfer roller 80 by a power unit, not shown. Further, arranged around the intermediate transfer body 50 is a corona charging device 52 functioning as an energy imparting means.

[0133]

The development apparatus 40 includes a development belt 41 functioning as a developer carrier, a black (hereinafter referred to as Bk) development unit 45K, a yellow (hereinafter referred to as Y) development unit 45Y, a magenta (hereinafter referred to as magenta) development unit 45M, and a cyan (hereinafter referred to as C) development unit 45C arranged around the development belt 41 in parallel with each other. In addition, the development belt 41 is suspended by a plurality of belt rollers and is arranged to be endlessly driven in a direction of an arrow by a drive means such as a motor, not shown. At a contact part on which the development belt 41 contacts the photoconductor 10, the development belt 41 moves at about a same speed as the photoconductor 10.

[0134]

Since each of the development units has a common configuration, a description is made only on a Bk development unit 50Bk. The other development unit 50Y, 50M, and 50C, are simply distinguished from the development unit 50Bk by suffixes Y, M and C attached to reference numerals corresponding to the numerals in the Bk development unit 50Bk. The development unit 50Bk includes a development tank 42Bk

for storing a liquid developer having a high viscosity and a high density, a scoop roller 43Bk arranged such that a lower part of the scoop roller 43Bk is immersed in the liquid developer in the development tank 42Bk, and an application roller 44Bk for applying the developer scooped up from the scoop roller 43Bk on the development belt 41 to form a thin layer. The application roller 44Bk is conductive and predetermined bias is applied on the application roller 44Bk from a power source, not shown.

[0135]

10 As for a configuration of the copier according to the present invention, in addition to the configuration illustrated in Fig. 1, the development unit 45 of each color may be arranged in parallel around the photoconductor 10 as illustrated in Fig. 2.

[0136]

15 Next, an operation of the copier according to the present invention is described. In Fig. 1, the photoconductor 10 is uniformly charged by the charging roller 20, wherein the photoconductor 10 is driven to rotate in a direction of an arrow. Then reflected light from an original document is subjected to image formation and projection by an optical system, not shown, of the exposure device 30 to form an electrostatic latent image on the photoconductor 10. The electrostatic latent image is developed by the development apparatus 40 and a toner image as a visible image is formed. The thin layer of developer on the development belt 41 is separated from the development belt 41 in a form of a thin layer by contact with the photoconductor in a development area. The separated thin layer is then transferred to a part on which the latent image on the photoconductor is formed. The toner image developed by the development apparatus 40 is transferred on a surface of the intermediate transfer body 50 in a contacting part (primary transfer area) between the photoconductor 10 and the intermediate transfer body 50 moving at a same speed as the photoconductor 10. When transfer is performed by superimposing

three or four colors, the above-mentioned process is repeated per each color to form a color image on the intermediate transfer body 50.

[0137]

5 The corona charging device 52 for applying charges on a superimposed toner image on the intermediate transfer body is arranged downstream in a part in which the photoconductor 10 and the intermediate transfer body 50 contacting and facing each other in a rotation direction of the intermediate transfer body 50, and
10 upstream in a location in which the intermediate transfer body 50 and the transfer paper 100 contacting and facing each other. The corona charging device 52 applies a true electric charge having a same polarity as a charging polarity of toner particles for forming the toner image to the toner image. Thus, the sufficient electric
15 charge is applied on the toner image to perform transfer on to the transfer paper in good condition. After the toner image is charged by the corona charging device 52, the toner image is entirely transferred (secondary transfer) on the transfer paper 100 conveyed to a direction of an arrow from a sheet feeding part, not shown,
20 by the transfer bias applied from the transfer roller 80. The transfer paper 100 on which the toner image is transferred is then separated from the photoconductor 10 by a separation device, not shown, and is discharged from the development apparatus after being subjected to fixing processing by a fixing device, now shown. On
25 the other hand, untransferred toner after the transfer is collected and removed from the photoconductor 10. Then residual charge is discharged by the discharge lamp 70 for next charging.

[0138]

As described above, a coefficient of static friction of the
30 intermediate transfer body is preferably 0.1 to 0.6, and more preferably 0.3 to 0.5. Volume resistance of the intermediate transfer body preferably is several Ω cm or more but less than 10^3 Ω cm. By setting the volume resistance to several Ω cm or more

but less than $10^3 \Omega \text{ cm}$, it prevents the charge applied by a charge imparting means from remaining on the intermediate transfer body as well as preventing the intermediate transfer body itself from taking charge. Accordingly, uneven transfer during the secondary transfer is prevented. In addition, the transfer bias may be easily applied during the secondary transfer.

[0139]

Materials for the intermediate transfer body are not specifically limited and any publicly known materials may be used. Examples of the materials for the intermediate transfer are as follows. (1) A material having a high Young's modulus (modulus of elasticity in tension) is used for a single-layer belt. Such materials include PC (polycarbonate), PVDF (polyvinylidene fluoride), PAT (polyalkylene terephthalate), a blended material of PC (polycarbonate)/PAT (polyalkylene terephthalate), a blended material of ETFE (ethylene-tetrafluoroethylene copolymer)/ PC, a blended material of ETFE/PAT, a blended material of PC/PAT, thermosetting polyimides containing dispersed carbon black, and so forth. The resulting single-layer belt having a high Young's modulus does not deform much stress applied during image formation. It is advantageous in that misregistration does not easily occur particularly during color image formation. (2) The intermediate transfer body may be a double or triple layer belt having a high Young's modulus as a base layer, wherein a surface layer or an intermediate layer is applied on a periphery of the base layer. The double or triple layer belt is capable of preventing a dropout of a line image caused by stiffness of a single layer belt. (3) The intermediate transfer body may also be a belt using a rubber or an elastomer having a relatively low Young's modulus. The belt is advantageous in that the dropout of the line image scarcely occurs due to its softness. In addition, by setting a width of the belt larger than the width of a drive roll or a suspension roll, the belt is prevented from meandering by using elasticity of an edge

part of the belt protruding from the roll. Accordingly low cost may be achieved without requiring a rib or a meandering prevention device.

[0140]

5 Conventionally, a flouororesin, a polycarbonate resin, and a polyimide resin are used for the intermediate transfer belt. Recently, a flexible belt, in which an entire layer of the belt or a part of the belt is formed of a flexible member, is used. Color image transfer using a resinous belt has following problems.

10 [0141]

Generally, a color image is formed by using toner of four colors. On one color image, one to four toner layers are formed. The toner layer is subjected to pressure by being undergoing the primary transfer (transfer from the photoconductor to the intermediate transfer belt) and the secondary transfer (transfer from the intermediate transfer belt to a sheet), resulting in high aggregation among toner. The higher the aggregation among toner, the easier a hollow defect in a character or a hollow defect in an edge of an image of a solid part occurs. Since the resinous belt having high hardness is resistant to deformation with respect to the toner layers, the toner layers may be easily compressed. Accordingly, a phenomenon of a hollow defect of a character may easily occur.

[0142]

25 Recently, there is a high demand in forming an image on various kinds of paper such as Japanese paper or embossed paper. However, paper having low smoothness often causes a gap between the toner and the paper during transfer, thereby toner dropout easily occurs. If a transfer pressure in a secondary transfer part is increased to improve adhesiveness, an aggregation force on the toner layer is increased. Accordingly, the above-described hollow defect of a character occurs.

[0143]

The flexible belt is used for following purposes. The flexible belt is deformed in the transfer part according to the toner layer and rough paper. In other words, since the flexible belt is deformed according to a local irregularity on paper, the transfer pressure applied on the toner layer may not be excessively increased. Accordingly, a transfer image with sufficient adhesiveness and uniformity without a hollow defect in a character may be achieved even on paper having poor planarity.

[0144]

Materials for the flexible belt include polycarbonate, a fluoro-resin (ETFE, PVDF), polystyrene, chloropolystyrene, poly- α -methylstyrene, styrene-butadiene copolymers, styrene-vinyl chloride copolymers, styrene-vinyl acetate copolymers, styrene-maleic acid copolymers, styrene-acrylate copolymers (styrene-methyl acrylate copolymers, styrene-ethyl acrylate copolymers, styrene-butyl acrylate copolymers, styrene-octyl acrylate copolymers, styrene-phenyl acrylate copolymers), styrene-methacrylate copolymers (styrene-methyl methacrylate copolymers, styrene-ethyl methacrylate copolymers, styrene-phenyl methacrylate copolymers), styrene-methyl α -chloroacrylate copolymers, a styrenic resin such as styrene-acrylonitrile-acrylate copolymers (homopolymers and copolymers containing styrene or a derivative substitution), a methyl methacrylate resin, a butyl methacrylate resin, an ethyl acrylate resin, a butyl acrylate resin, a modified acrylic resin (a silicone-modified acrylic resin, a vinyl-chloride-modified acrylic resin, and an acrylic-urethane resin), a vinyl chloride resin, styrene-vinyl acetate copolymers, vinyl chloride-vinyl acetate copolymers, a rosin-modified maleic acid resin, a phenolic resin, an epoxy resin, a polyester resin, a polyester polyurethane resin, polyethylenes, polypropylenes, polybutadienes, polyvinylidene chloride, an ionomer resin, a polyurethane resin, a silicone resin, a ketone resin, ethylene-ethyl acrylate

copolymers, a xylene resin, a polyvinyl butyral resin, a polyamide resin, a modified polyphenylene oxide resin, and so forth. Each of these resins may be used alone or in combination with two or more kinds of resins. However, it is to be noted that the material of the flexible belt is obviously not limited to the above-described materials.

[0145]

Materials for the flexible belt or the elastomer includes butyl rubber, fluorocarbon rubber, acrylic rubber, EPDM, NBR, acrylonitrile-butadiene-styrene rubber, natural rubber, isoprene rubber, styrene-butadiene rubber, butadiene rubber, ethylene-propylene rubber, ethylene-propylene terpolymers, chloroprene rubber, chlorosulfonated polyethylenes, chlorinated polyethylenes, urethane rubber, syndiotactic 1,2-polybutadiene, epichlorohydrin rubber, silicone rubber, fluorocarbon rubber, polysulfide rubber, polynorbornene rubber, hydrogenated nitrile rubber, thermoplastic elastomers (polystyrene elastomers, polyolefin elastomers, polyvinyl chloride elastomers, polyurethane elastomers, polyamide elastomers, polyurea elastomers, polyester elastomers, fluoroelastomers), and so forth. Each of these materials may be used alone or in combination with two or more kinds of materials. However, it is to be noted that the materials of the flexible belt is obviously not limited to the above-described materials.

[0146]

A conducting agent for adjusting a resistance value is not specifically limited. For instance, carbon black, graphite, metal powder such as aluminum powder and nickel powder, conductive metal oxides such as tin oxide, titanium oxide, antimony oxide, indium oxide, potassium titanate, antimony-tin complex oxide (ATO), indium-tin complex oxide (ITO), and so forth, may be covered with insulative fine particles of barium sulfate, magnesium silicate, calcium carbonate, and so forth. It is to be noted that the

conducting agent is obviously not limited to the above-described conductive agent.

[0147]

The surface layer of the flexible belt and a material of the surface layer are required to prevent contamination on the photoconductor caused by an elastic material, and to reduce an adhesion force of the toner by reducing surface frictional resistance on the surface of the transfer belt to improve cleaning performance and the secondary transfer. For instance, the material of the surface layer may be formed of polyurethane, polyester, and an epoxy resin. The materials may be used alone or in combination with two or more kinds of materials to reduce the surface energy. A material for increasing a lubricating characteristic such as powders and particles of fluororesins, fluorine compounds, carbon fluoride, titanium dioxide, silicon carbide, and so forth, may be dispersed in the material of the surface layer, and they may be used alone or in combination with two or more kinds, or may have different particle diameters. In addition, a material such as fluorine rubber which is subjected to heat processing and thereby form a layer rich in fluorine on the surface and reduce the surface energy may be used.

[0148]

A production method of the belt is not limited. For instance, a centrifugal molding method in which materials are placed in a rotating cylindrical mold to form a belt, a spray coating method in which a liquid coating composition is sprayed to form a film, a dipping method in which a cylindrical mold is dipped in a solution of the material and is then taken out, an injection method in which a material composition is injected into an inner mold or an outer mold, and a method in which a compound is placed around a cylindrical mold and is subjected to vulcanization and polishing may be included. The production method is not limited to the above-described methods. A plurality of production methods are generally used in combination

to produce a belt.

[0149]

A method of preventing the flexible belt from elongation includes a method of forming a rubber layer on a core resin layer having less elongation, a method of incorporating a material in a core layer for preventing the elongation, and so forth. However, these methods are not specifically relating to the production method of the flexible belt. Materials for forming the core layer for preventing elongation include, but are not limited to, natural fibers such as cotton, silk, and so forth, synthetic fibers such as polyester fibers, nylon fibers, acrylic fibers, polyolefin fibers, polyvinyl alcohol fibers, polyvinyl chloride fibers, polyvinylidene chloride fibers, polyurethane fibers, polyacetal fibers, polyfluoroethylene fibers, phenol fibers, and so forth, inorganic fibers such as carbon fibers, glass fibers, and boron fibers, metallic fibers such as iron fibers and copper fibers. A woven material or a thread is fabricated by selectively using one or more than two kinds of the above-mentioned materials.

[0150]

The thread may take any form of threads including a thread formed of plying one or a plurality of filaments, a single twist thread, a plied thread, a two ply thread, and so forth. In addition, materials selected from the above-mentioned materials may be blended to form a fiber. Appropriate conduction processing may naturally be performed on the thread before using the thread.

[0151]

On the other hand, any woven material such as a knitted fabric may be used. A looped fabric may naturally be used and the conduction processing may naturally be performed on the looped fabric. A method of forming the core layer is not limited but includes a method of placing a cylindrically woven fabric on a mold, and so forth, and form a coating layer on the cylindrically woven fabric, a method of dipping the cylindrically woven fabric in a

liquid rubber and thereby forming a coating layer on one side or both sides of the core layer, a method of spirally placing a thread around a mold, and so forth, at an optional pitch and forming a coating layer on the thread, and so forth.

5 Thickness of a flexible layer depends on hardness of the flexible layer. However, when the thickness of the flexible layer is too thick, elongation of the surface of the flexible layer increases and a cracking on the surface may easily occur. Further, when the elongation increases, an image may be subjected to elongation and
10 contraction, and so forth. Therefore, it is not preferable to have a thick flexible layer (approximately 1 mm or more).

[0152]

(Tandem type color image forming apparatus)

The present invention may be used in the tandem type color image
15 forming apparatus. One embodiment of the tandem type color image forming apparatus is described below. There are two types of tandem type color image forming apparatus; one using a direct transfer method of sequentially transferring an image on each of the photoconductors 1 to a sheet s which is conveyed by a conveyance
20 belt 3 by a transfer device 2, and the other using an indirect transfer method of temporarily and sequentially transferring an image on each of the photoconductors 1 on an intermediate transfer body 4 by a primary transfer device 2, and then transferring the image on the intermediate transfer body 4 to the sheet s all at
25 once by a secondary transfer device 5. The transfer device 5 in the embodiment is a transfer conveyance belt, however, a belt in a form of a roller may be used. The direct transfer method and the indirect transfer method are illustrated in Fig. 3 and Fig. 4, respectively.

30 [0153]

Comparison between the direct transfer method and the indirect transfer method is made. In the direct transfer method, a sheet feeding device 6 is to be provided upstream of a tandem type image

forming apparatus T in which photoconductors 1 are arranged in parallel, and a fixing device 7 is to be provided downstream of the tandem type image forming apparatus T. Accordingly, the tandem type image forming apparatus T using the direct transfer method is disadvantageous in that a size of the apparatus is increased in a sheet conveying direction.

In contrast, in the indirect transfer method, a secondary transferring position may be relatively freely arranged.

Further, the sheet feeding device 6 and the fixing device 7 may be arranged above or below the tandem type image forming apparatus T. Accordingly, the tandem type image forming apparatus T using the indirect transfer method is advantageous in that the apparatus may be downsized.

[0154]

In addition, in the direct transfer method, the fixing device 7 is arranged in a vicinity of the tandem type image forming apparatus T to avoid increasing the size of the apparatus in the sheet conveying direction. For this reason, the fixing device 7 may not be arranged with sufficient space allowing the sheet s to bend. Accordingly, image formation upstream of the image fixing device 7 is easily affected by an impact caused when a tip of the sheet s enters the fixing device 7 (particularly significant when using a thick sheet) and by a difference in speed between a sheet conveying speed when the sheet s passes through the fixing device 7 and a sheet conveying speed of the transfer conveyance belt.

[0155]

On the other hand, in the indirect transfer method, the fixing device 7 may be arranged with a sufficient space allowing the sheet s to bend. Accordingly, the fixing device 7 may be arranged so as not to significantly affect image formation.

[0156]

For reasons as described above, recently, tandem type electrophotographic apparatus, particularly using an indirect

transfer method, is a focus of attention.

[0157]

In addition, as illustrated in Fig. 4, in color electrophotographic apparatus of the kind, a surface of the photoconductor 1 is cleaned by removing residual toner remained on the photoconductor 1 after the primary transfer by a photoconductor cleaning device 8 for next image formation. In addition, a surface of the intermediate transfer body 4 is cleaned by removing residual toner remained on the intermediate transfer body 4 by the intermediate transfer body

[0158]

An embodiment using the toner and the developer according to the present invention is described below with reference to figures.

[0159]

Fig. 5 illustrates one embodiment using the toner and the developer according to the present invention. Reference numerals 100, 200, 300, and 400 in Fig. 5 indicate a copier body, a sheet feeding table on which the copier 100 is mounted, a scanner arranged on the copier 100, and an automatic document feeder (ADF) arranged further on top of the scanner 300, respectively. An intermediate transfer body 10 in a form of an endless belt is provided in a center of the copier 100.

[0160]

In addition, as illustrated in Fig. 5, the intermediate transfer body 10 is spanned around three support rollers 14, 15, and 16, and may be rotated or conveyed clockwise as illustrated in Fig. 5.

[0161]

In the embodiment in Fig. 5, an intermediate transfer body cleaning device 17 for removing residual toner remained on the intermediate transfer body 10 after image transfer is provided on a left side of the support roller 15, which is a second support roller among three support rollers.

[0162]

In addition, above the intermediate transfer body 10 spanned between a first support roller 14 and the second support roller 15 among three support rollers, four image formation means for colors of yellow, cyan, magenta, and black are arranged side by side in a moving direction of the intermediate transfer body 10, forming tandem type image forming apparatus 20.

[0163]

As illustrated in Fig. 5, above the tandem type image forming apparatus 20, an exposure device 21 is further arranged. On the other hand, a secondary transfer device 22 is arranged on an opposite side of the tandem type image forming apparatus 20 across the intermediate transfer body 10. In an example illustrated in Fig. 5, the secondary transfer belt 24 which is the endless belt is spanned around two rollers 23. The secondary transfer belt 24 is arranged such that the secondary transfer belt 24 is pressed on a third support roller through the intermediate transfer body 10. Thus an image on the intermediate transfer body 10 is transferred on a sheet.

[0164]

Beside the secondary transfer device 22, a fixing device 25 for fixing a transfer image on the sheet is provided. The fixing device 25 is formed of a fixing belt 26, which is an endless belt, and a pressing roller 27 pressing the fixing belt 26.

[0165]

The above-described secondary transfer device 22 also includes a sheet conveyance function for conveying a sheet after being subjected to image transfer to the fixing device 25. Naturally, a transfer roller or a non-contact charger may be used as the secondary transfer device 22. In such a case, it becomes difficult to additionally provide the sheet conveyance function in the secondary transfer device 22.

[0166]

In the embodiment in Fig. 5, a sheet reversing device 28 is

arranged below the secondary transfer device 22 and the fixing device 25, in parallel with the above-described tandem type image forming apparatus 20. The sheet reversing device 28 reverses a sheet so that images are recorded on both sides of the sheet.

5 [0167]

When making a copy by using the color electrophotographic apparatus, set a document on a manuscript table 30 of the automatic document feeder 400. Alternatively, the document may be set on a contact glass 32 of the scanner 300 by opening the automatic document feeder 400, and then close the automatic document feeder 400 so as to hold the document.

[0168]

When the document is set on the automatic document feeder 400 and a start switch, not shown, is pressed, the document is conveyed and transferred onto the contact glass 32, and then a first carriage 33 and a second carriage 34 are driven. Alternatively, when the document is set on the contact glass 32, the scanner 300 is immediately driven to drive the first carriage 33 and the second carriage 34. Light is emitted from a light source at the first carriage 33 and reflected light from a surface of the document is further reflected toward the second carriage 34. The reflected light is then further reflected by a mirror of the second carriage 34 so as to pass through an image-forming lens 35 into a reading sensor 36 to read a content of the document, accordingly.

25 [0169]

In addition, when the start switch, not shown, is pressed, a drive motor, not shown, rotates and drives one of the support rollers 14, 15, and 16 to thereby cause the remaining two support rollers to rotate followed by the rotation of the one support roller to rotatively convey the intermediate transfer body 10. At the same time, the photoconductors 40 is rotated by the respective image forming means 18 to thereby form black, yellow, magenta, and cyan monochrome images on the photoconductors 40, respectively. Along

with the conveyance of the intermediate transfer body 10, the monochrome images are sequentially transferred to form a composite color image on the intermediate transfer body 10.

[0170]

5 On the other hand, when the start switch, not shown, is pressed, one of feeder rollers of a feeder table 200 is selectively rotated, sheets are ejected from one of multiple sheet feeding cassettes 44 in a paper bank 43, and the sheets are separated one by one by a separation roller 45. The separated sheet is placed in a sheet
10 feeding path, conveyed by a conveyance roller 47 into a sheet feeding path 48 in a copier 100, and caused to stop by colliding with a resist roller 49.

[0171]

Alternatively, when the start switch, not shown, is pressed,
15 sheets on a manual insertion tray 51 are ejected by rotating a sheet feeding roller 50 and the sheets are separated one by one by a separation roller 52. The separated sheet is placed in a manual insertion path 53 and similarly caused to stop by colliding with the resist roller 49.

20 [0172]

Then the resist roller 49 is rotated in exact timing with formation of the composite color image on the intermediate transfer body 10 to feed a sheet between the intermediate transfer body 10 and the secondary transfer device 22, and the composite color image
25 is transferred on the sheet by the secondary transfer device 22 to record a color image.

[0173]

The sheet after being subjected to image transfer is conveyed by the secondary transfer device 22 into the fixing device 25 and
30 heat and pressure are applied by the fixing device 25 to fix the transferred image. Then the sheet is ejected by an ejection roller 56 by changing a conveyance direction by a switch blade 55 and stacked on a sheet ejection tray 57. Alternatively, the sheet may be placed

in the sheet reversing device 28 by changing the conveyance direction by the switch blade 55, reversed, and again fed to a transfer position. After an image is recorded also on a back side of the sheet, the sheet is discharged on the sheet ejection tray
5 57 by the ejection roller 56.

[0174]

On the other hand, in the intermediate transfer body after the image transfer, residual toner remained on the intermediate transfer body 10 after the image transfer is removed by the
10 intermediate transfer cleaning device 17 for next image formation.

[0175]

The resist roller 49 is generally earthed, however, bias may be applied to remove paper dust on the sheet.

[0176]

15 As illustrated in Fig. 6 in detail, each of the image forming means 18 in the tandem image forming apparatus 20 includes a charging device 60, development apparatus 61, a primary transfer device 62, a photoconductor cleaning device 63, and a discharging device 64 around the drum-shape photoconductor 40.

20 [0177]

[Embodiment]

The present invention is concretely described below with reference to the embodiment and a comparative example. However, use of the present invention is not limited to the embodiment and
25 the comparative example. In addition, a part and % in a following example represent an amount by weight, unless otherwise specified. A test machine used in the embodiment and an obtained characteristic and a result are shown in Table 1. In the embodiment, evaluation is carried out as follows.

30 [0178]

(Test machine)

One of following test machines A, B, C, and D is used to evaluate an image used in the evaluation.

(Test machine A)

The test machine A for use in the evaluation is a modified tuned tandem type full-color laser printer PISiO Color 800 available from Ricoh Company, Ltd., including a four-color non-magnetic two-component development part and photoconductors for four colors in which an original image fixing unit is improved to be an oilless fixing unit. The evaluation of a printing speed is carried out in a high-speed printing mode (variation from 20 to 50 A4-sized sheets/min.).

[0179]

(Test machine B)

The test machine B for use in the evaluation is a modified tandem type full-color laser printer PISiO Color 800 available from Ricoh Company, Ltd., including a four-color non-magnetic two-component development part and photoconductors for four colors in which an original transfer system is modified to an intermediate transfer system for primarily transferring a toner image on an intermediate transfer body and secondarily transferring the toner image on a transfer material, and an original fixing unit is improved and tuned to be an oilless fixing unit. The evaluation of a printing speed is carried out in a high-speed printing mode (variation from 20 to 50 A4-sized sheets/min.).

[0180]

(Test machine C)

The test machine for use in the evaluation is a modification of a full-color laser copier IMAGIO Color 2800 available from Ricoh Company, Ltd. of which an original fixing unit is improved and tuned to be an oilless fixing unit. In the original copier, four-color development parts develop four color images on one drum-shaped photoconductor using a two-component developer, the four color images are sequentially transferred on an intermediate transfer body, and the four color images are then transferred on a transfer material all at once.

[0181]

(Test machine D)

The test machine D for use in the evaluation is a modification of a full-color laser printer IPSiO Color 5000 available from Ricoh Company Ltd., of which an original fixing unit is improved and tuned to be an oilless fixing unit but the printer is tuned as an oil-applied type printer. In the original printer, four-color development parts sequentially develop four-color images on one belt photoconductor using non-magnetic one-component developer, the four color images are sequentially transferred on an intermediate transfer body, and the four color images are then transferred on a transfer material all at once.

[0182]

(Test machine E)

The test machine E for use in the evaluation is a modification of a tandem type full-color laser printer IPSiO Color 8000 available from Ricoh Company, Ltd., including a four-color non-magnetic two-component development part and four-color photoconductors, of which an original oil-coated fixing part is used as intact. The evaluation of a printing speed is carried out in a high-speed printing mode (variation from 20 to 50 A4-sized sheets/min.).

[0183]

(Evaluation item)

1) Burying characteristic of an additive agent

A burying condition of an external additive is performed as follows. A sample toner is stored at 40 °C and 80% humidity for one week, is then agitated in a development unit for one hour, and a surface of the toner is observed by using a FE-SEM (field emission scanning electron microscope S-4200 available from Hitachi, Ltd.). The less the burying, the better. The burying of the external additive is rated as X, Δ , O, and \odot , in order of ascending rank.

[0184]

2) Toner dispersibility

After continuously outputting 30,000 copies of an image chart in a monochrome mode with an image area of 50%, a the development unit is opened and an amount of toner scattered from a development part is visually determined and is rated as X, Δ, O, and ⊙, in order of ascending rank.

[0185]

3) Hollow defect in character image

After continuously outputting 30,000 copies of an image chart in a monochrome mode with an image area of 50%, four color character images are superimposedly output on an OHP sheet of a DX type available from Ricoh Company, Ltd. And a frequency of untransferred toner in which a line image of a character part is hollowed is compared to a stepwise quality grade sample, and is rated as X, Δ, O, and ⊙, in order of ascending rank.

[0186]

4) Toner transfer rate

After continuously outputting 200,000 copies of an image chart in a monochrome mode with an image area of 7%, the transfer rate is calculated based on relation between an amount of supplied toner and an amount of wasted toner.

Transfer rate (%) = $100 \times (\text{amount of supplied toner} - \text{amount of wasted toner}) / (\text{amount of supplied toner})$

⊙, O, Δ, and × indicate the transfer rate of 90 or more, the transfer rate of 75 or more but less than 90, the transfer rate of 60 or more but less than 75, and the transfer rate of less than 60, respectively.

[0187]

5) Toner supply characteristic

An image chart with an image area of 90% and an image chart with an image area of 5% are alternately output every 4000 sheets. The toner supply characteristic at the time is examined, and is rated as X, Δ, O, and ⊙, in order of increasing supply characteristic.

[0188]

6) Transfer dust

After continuously outputting 30,000 copies of an image chart in a monochrome mode with an image area of 50%, each of four-color solid images of 10 mm X 10 mm is superimposedly output on Type 6000 paper available from Ricoh Company, Ltd., and the transfer dust is compared to a stepwise quality grade sample, and is rated as X, Δ , O, and \odot , in order of ascending rank.

[0189]

7) Thin line reproducibility

After continuously outputting 30,000 copies of an image chart in a monochrome mode with an image area of 50%, an image of a thin line of 600 dpi is output on Type 6000 Paper available from Ricoh Company, Ltd. Blurring of the thin line is compared to a stepwise quality grade sample, and is rated as X, Δ , O, and \odot , in order of ascending rank.

[0190]

8) Background soiling

After continuously outputting 30,000 copies of an image chart in a monochrome mode with an image area of 50%, the test machine is stopped in a course of development of a blank image. A developer on a photoconductor after the development is transferred onto a tape. A difference in image density between the tape on which the developer is transferred and a tape on which the developer is not transferred is determined by using a Model 938 spectrodensitometer (available from X-Rite, Inc.). The smaller the difference in the image density is, the less the background soiling is. The background soiling is rated as X, Δ , O, and \odot , in order of ascending rank.

[0191]

9) Image density

After continuously outputting 150,000 copies of an image chart in a monochrome mode with an image area of 50%, a solid image is output on Type 6000 paper available from Ricoh Company, Ltd. Then

the image density of the solid image is determined by using an X-Rite (available from X-Rite, Inc.). This procedure is performed on four colors, respectively, and an average density of four colors is determined. ×, Δ, ○, and ◎ indicates the average value of less than 1.2, the average value of 1.2 or more but less than 1.4, the average value of 1.4 or more but less than 1.8, and the average value of 1.8 or more but less than 2.2, respectively.

[0192]

10) Thermal storage stability

10 A total of 10 g of each color toner is weighed and is placed in a 20-ml glass vessel, the glass vessel is then tapped for 100 times and is left to stand in a thermostat chamber set at 55 °C for 24 hours. Then a needle penetration degree of a sample is determined by a penetrometer. The needle penetration degree is indicated by
15 ◎: 20 mm or more, ○: 15 mm or more but less than 20 mm, Δ: 10 mm or more but less than 15 mm, and ×: less than 10 mm, in order of favorable thermal storage stability.

[0193]

11) Transparency

20 After continuously outputting 100,000 copies of an image chart in a monochrome mode with an image area of 50%, images of each color are fixed on an OHP sheet Type DX available from Ricoh Company, Ltd. under a condition of an image density of 1.0 mg/cm² and at a fixing temperature of 140 °C. The transparency of the fixed image
25 is determined by a direct reading Haze Computer Model HGM-2DP available from Suga Test Instruments Co., Ltd., and the transparency is rated as ◎, ○, Δ, and ×, in order of favorable transparency.

[0194]

14) Clearness and reproducibility of color

30 After continuously outputting 100,000 copies of an image chart in a monochrome mode with an image area of 50%, an image is output on 6000 Paper available from Ricoh Company, Ltd. The clearness and reproducibility of color of the image are visually evaluated and

are rated as ◎, ○, △, and ×, in order of favorable clearness and reproducibility of the color.

[0195]

13) Glossiness

5 After continuously outputting 100,000 copies of an image chart in a monochrome mode with an image area of 50% on 6000 Paper available from Ricoh Company, Ltd., the glossiness of the images is determined by using a gloss meter VG-1D available from Nippon Denshoku Industries, Co., Ltd. at a transmission angle of 60° and an
10 acceptance angle of 60° with an S mode in an S-S/10 switch after zero point adjustment and standard setting using a standard plate are performed. The glossiness is indicated by ◎: 15 mm or more, ○: 6 mm or more but less than 15 mm, △: 3 mm or more but less than 6 mm, and ×: less than 3 mm, in order of favorable glossiness.

15 [0196]

14) Charging stability under a high-temperature and high-humidity environment

While continuously outputting 100,000 copies of an image chart in a monochrome mode with an image area of 7% at a temperature of
20 40° and a humidity of 90%, a part of a tested developer is sampled every 1000 copies. An amount of charges of the sampled developer is determined according to a blow-off method, and the charging stability is rated as ◎, ○, △, and ×, in order of favorable charging stability with less decrease in charging.

25 [0197]

15) Charging stability under a low-temperature and low-humidity environment

While continuously outputting 100,000 copies of an image chart in a monochrome mode with an image area of 7% at a temperature of
30 10° and a humidity of 15%, a part of a tested developer is sampled every 1000 copies. An amount of charges of the sampled developer is determined according to a blow-off method, and the charging

stability is rated as ⊙, ○, △, and ×, in order of favorable charging stability with less decrease in charging.

[0198]

16) Fixing characteristic

5 A minimum fixing temperature and a maximum fixing temperature are adequately set in a fixing temperature range. Accordingly, hot offset and cold offset does not occur and transportation problems such as wraparound and paper jamming hardly occur. Overall fixing characteristic is evaluated and is rated as ⊙, ○, △, and ×, in
10 order of favorable fixing characteristic.

[0199]

(Evaluation on two-component developer)

The two-component developer for use in evaluating images is prepared by uniformly mixing 5 parts by weight of each color toner
15 with 100 parts by weight of a carrier in a tumbler mixer, in which a container is tumbled to agitate the contents, and charging the resulting mixture. A ferrite carrier having an average particle diameter of 50 μm coated with a silicone resin having an average thickness of 0.3 μm is used.

20 [0200]

(Production of carrier)

• Core material

Cu-Zn ferrite particles (weight-average particle diameter: 35 μm) 5000 parts

25 • Coating material

Toluene 450 parts

Silicone resin SR 2400 (available from Dow Corning Toray Silicone Co., Ltd.; nonvolatile content 50%) 450 parts

Aminosilane SH 6020 (available from Dow Corning Toray Silicone Co.,
30 Ltd.) 10 parts

Carbon black 10 parts

The coating material is dispersed for 10 minutes by a stirrer to

adjust a coating liquid. The coating liquid and the core material are placed in a coating device for coating. In the coating device, a rotary base plate disk and an agitator impellor are provided on a fluidized bed, and coating is performed while a revolving current is formed. Thus the coating liquid is applied on the core material. An obtained coated material is then burned in an electric oven at 250 °C for 2 hours to obtain the carrier.

[0201]

Embodiment 1

(Oxide fine particles 1)

Methyltrimethoxysilane purified by distillation is heated, followed by bubbling of nitrogen gas into the heated methyltrimethoxysilane. The methyltrimethoxysilane is then introduced into an oxyhydrogen flame burner with flow of nitrogen gas and is burnt and decomposed in the oxyhydrogen flame. An added amount of methyltrimethoxysilane, an oxygen gas, a hydrogen gas, and a nitrogen gas are 1270 g/hr, 2.9 Nm³/hr, 2.1 Nm³/hr, and 0.58 Nm³/hr, respectively. Generated spherical silica fine powder is collected by a bag filter. 1 kg of the spherical silica fine powder is placed in a 5-liter planetary mixer, and 10 g of pure water is added to the spherical silica fine powder with stirring. After sealing the mixer, the mixture is stirred at 55 °C for 14 hours. After cooling to room temperature, 20 g of hexamethyldisilazane is added in the mixture by agitation. After sealing the mixer, the mixture is agitated for further 24 hours. The temperature in the mixture rises to 115 °C. A remained material and produced ammonia are removed under flow of nitrogen gas. Thus the oxide fine particles 1 are obtained. A primary particle diameter and circularities SF1 and SF2, and uniformity of elementary composition of the obtained oxide fine particles 1 are shown in Table 2.

[0202]

(Oxide fine particles 2)

Oxide fine particles 2 are prepared by the same procedure as the

oxide fine particles 1, except that the amounts of methyltrimethoxysilane, oxygen gas, and hydrogen gas are changed to 880 g/hr, 2.3 Nm³/hr, and 1.8 Nm³/hr, respectively. The primary particle diameter, the standard deviation thereof, circularities, and uniformity of elementary composition of the obtained oxide fine particles 2 are shown in Table 2.

(Oxide fine particles 3)

Oxide fine particles 3 are prepared by the same procedure as the oxide fine particles 1, except that the amounts of methyltrimethoxysilane, oxygen gas, and hydrogen gas are changed to 1430 g/hr, 3.1 Nm³/hr, and 2.3 Nm³/hr, respectively. The primary particle diameter, the standard deviation thereof, circularities, and uniformity of elementary composition of the obtained oxide fine particles 3 are shown in Table 2.

(Oxide fine particles 4)

Oxide fine particles 4 are prepared by the same procedure as the oxide fine particles 1, except that the amounts of methyltrimethoxysilane, oxygen gas, and hydrogen gas are changed to 1520 g/hr, 3.4 Nm³/hr, and 2.5 Nm³/hr, respectively. The primary particle diameter, the standard deviation thereof, circularities, and uniformity of elementary composition of the obtained oxide fine particles 4 are shown in Table 2.

[0203]

(Oxide fine particles 5)

Methyltrimethoxysilane purified by distillation is heated, followed by bubbling of a nitrogen gas into the heated methyltrimethoxysilane. The methyltrimethoxysilane and titanium (Ti) metallic particles are then introduced into an oxyhydrogen flame burner with flow of nitrogen gas and is burnt and decomposed in the oxyhydrogen flame. An added amount of the methyltrimethoxysilane, titanium metallic particles, an oxygen gas, a hydrogen gas, and a nitrogen gas are 1270 g/hr, 127 g/hr, 2.9 Nm³/hr, 2.1 Nm³/hr, and 0.58 Nm³/hr, respectively. Generated

spherical silica fine powder containing Ti is collected by a bag filter. 1 kg of the spherical silica fine powder is placed in a 5-liter planetary mixer, and 10 g of pure water is added to the spherical silica fine powder with stirring. After sealing the mixer, the mixture is stirred at 55 °C for 14 hours. After cooling to room temperature, 20 g of hexamethyldisilazane is added in the mixture by agitation. After sealing the mixer, the mixture is agitated for further 24 hours. The temperature in the mixture rises to 115 °C. A remained material and produced ammonia are removed under flow of a nitrogen gas. Thus the oxide fine particles 1 are obtained. A primary particle diameter and circularities SF1 and SF2, and uniformity of elementary composition of the obtained oxide fine particles 1 are shown in Table 2.

[0204]

(Oxide fine particles 6)

Oxide fine particles 6 are prepared by the same procedure as the oxide fine particles 5, except that zinc is used instead of titanium. The primary particle diameter, circularities SF1 and SF2, and uniformity of elementary composition of the obtained oxide fine particles 6 are shown in Table 2.

[0205]

(Oxide fine particles 7)

Oxide fine particles 7 are prepared by the procedure of the oxide fine particles 1, except that 100 g of the prepared oxide fine particles is dispersed in 400 g of toluene, 5 g of dimethylsilicone oil having a viscosity of 300 cs is added in the toluene mixture, and the toluene is removed by heating and distillation. The primary particle diameter, circularities SF1 and SF2, and uniformity of elementary composition of the obtained oxide fine particles 7 are shown in Table 2. A liberation degree of the silicone oil in the oxide fine particles is 75%.

[0206]

(Oxide fine particles 8)

Methyltrimethoxysilane purified by distillation is heated, followed by bubbling of nitrogen gas into the heated methyltrimethoxysilane. The methyltrimethoxysilane is then introduced into an oxyhydrogen flame burner with flow of a nitrogen gas and is burnt and decomposed in the oxyhydrogen flame. An added amount of methyltrimethoxysilane, an oxygen gas, a hydrogen gas, and a nitrogen gas are 980 g/hr, 3.3 Nm³/hr, 5.0 Nm³/hr, and 1.23 Nm³/hr, respectively. Generated spherical silica fine powder is collected by a bag filter. 1 kg of the spherical silica fine powder is placed in a 5-liter planetary mixer, and 10 g of pure water is added to the spherical silica fine powder with stirring. After sealing the mixer, the mixture is stirred at 55 °C for 14 hours. After cooling to room temperature, 20 g of hexamethyldisilazane is added in the mixture by agitation. After sealing the mixer, the mixture is agitated for further 24 hours. The temperature in the mixture rises to 115 °C. A remained material and produced ammonia are removed under flow of a nitrogen gas. Thus the oxide fine particles 8 are obtained. A primary particle diameter and circularities SF1 and SF2, and uniformity of elementary composition of the obtained oxide fine particles 1 are shown in Table 2.

[0207]

(Oxide fine particles 9)

Silicon tetrachloride is burnt and hydrolyzed in oxyhydrogen flames to obtain silica, and the silica is deposited and concurrently melted on a heat-resistant carrier and thereby obtain silica fine particles. Porous glass base material is produced from the silica fine particles and the porous glass base material is melted to obtain the oxide fine particles 9. Primary particle diameter, circularities SF1 and SF2, and uniformity of elementary composition of the obtained oxide fine particles 9 are shown in Table 2.

[0208]

(Oxide fine particles 10)

An alkoxysilane is hydrolyzed in an alcohol (ethanol) solvent in the presence of an acidic catalyst to obtain a silica sol. The silica sol is converted into gel, dried, calcined, and sintered to obtain oxide fine particles 10. Primary particle diameter, circularities SF1 and SF2, and uniformity of elementary composition of the oxide fine particles 10 are shown in Table 2.

[0209]

(Oxide fine particles 11)

Methyltrimethoxysilane purified by distillation is heated, followed by bubbling of nitrogen gas into the heated methyltrimethoxysilane. The methyltrimethoxysilane is then introduced into an oxyhydrogen flame burner with flow of nitrogen gas and is burnt and decomposed in the oxyhydrogen flame. An added amount of methyltrimethoxysilane, oxygen gas, hydrogen gas, and nitrogen gas are 1270 g/hr, 1.1 Nm³/hr, 0.9 Nm³/hr, and 1.2 Nm³/hr, respectively. Generated spherical silica fine powder is collected by a bag filter. 1 kg of the spherical silica fine powder is placed in a 5-liter planetary mixer, and 10 g of pure water is added to the spherical silica fine powder with stirring. After sealing the mixer, the mixture is stirred at 55 °C for 1 hour. After cooling to room temperature, 20 g of hexamethyldisilazane is added in the mixture by agitation. After sealing the mixer, the mixture is agitated for further 8 hours. The temperature in the mixture rises to 115 °C. A remained material and produced ammonia are removed under flow of nitrogen gas. Thus the oxide fine particles 1 are obtained. A primary particle diameter and circularities SF1 and SF2, and uniformity of elementary composition of the obtained oxide fine particles 1 are shown in Table 2.

[0210]

(Polyol resin 1)

378.4 g of a low-molecular-weight bisphenol A epoxy resin (number-average molecular weight: about 360), 86.0 g of a high-molecular-weight bisphenol A epoxy resin (number-average

molecular weight: about 2700), 191.0 g of a glycidylated adduct of bisphenol A propylene oxide ($n + m$ is about 2.1 in the Formula (1)), 274.5 g of bisphenol F, 70.1 g of p-cumylphenol, and 200 g of xylene are added in a separable flask with a stirrer, a thermometer, an N₂ inlet, and a cooling tube. The resulting mixture is heated to a temperature of 70 to 100 °C in an atmosphere of N₂ and 0.183 g of lithium chloride is added, and is further heated to a temperature of 160 °C. Water is then added to the mixture under reduced pressure and is bubbled together with xylene to remove water, xylene, other volatile components, and polar-solvent-soluble matters. The residue is caused to react at 180 °C for 6 to 9 hours for polymerization and thereby obtained 1000 g of polyol resin (Polyol Resin 1) with Mn : 3800, Mw/Mn : 3.9, Mp : 5000, a softening point of 109 °C, Tg 58 °C, and a weight per epoxy equivalent of 20000 or more. In the polymerization reaction, reaction conditions are controlled so that monomer components does not remain. A polyoxyalkylene moiety in a main chain is identified by NMR.

[0211]

(Production of toner)

Black toner

Water	1000 parts
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Phthalocyanine Green hydrous cake (solid content 30%)	200 parts
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Carbon black (MA 60, available from Mitsubishi Chemical Corp.)	540 parts
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Polyol resin 1	1200 parts
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The above materials are mixed in a Henschel mixer and a mixture in which pigment aggregates are impregnated with water is obtained. The mixture is kneaded in a twin-roll mill of which a roll surface temperature is set to 130 °C for 45 minutes, rolled and cooled, and pulverized by a pulverizer to obtain a master batch pigment.

[0212]

Polyol resin 1	100 parts
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Master batch	8 parts
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Charge control agent (BONTRON E-84,
available from Orient Chemical Industries, Ltd.) 2 parts
Wax (fatty acid ester wax, melting point: 83 °C, viscosity: 280
mPa •s (90 °C)) 5 parts

5 The above material is mixed in a mixer, is then melted and kneaded
in a two-roll mill for three or more times, and the kneaded material
is rolled and cooled. The resulting material is then pulverized
in a pulverizer (I-Type Mill : available from Nippon Pneumatic MFG.
Co., Ltd.) of a jet mill breaker disc system, and subjected to air
10 classification by action of a revolving current (DS classifier :
available from Nippon Pneumatic MFG. Co., Ltd.) and thus black
particles having a volume-average particle diameter of 5.5 μm is
obtained. The black particles are further mixed with 3.0% by weight
of the oxide fine particles 1 and 1.0% by weight of a hydrophobic
15 silica (HDK H 2000, available from Clariant Japan K.K.) having a
primary particle diameter of 10 nm in a Henschel mixer, the resulting
mixture is passed through a sieve with an aperture of 50 μm to remove
aggregates. The black toner 1 is thus obtained. A dispersion
diameter of the wax in the toner is 0.5 μm .

20 [0213]

Yellow toner

Water 600 parts

Pigment Yellow 17 hydrous cake (solid content 50%) 1200 parts

Polyol resin 1 1200 parts

25 The above materials are mixed in a Henschel mixer and a mixture
in which pigment aggregates are impregnated with water is obtained.
The mixture is kneaded in a twin-roll mill, of which a roll surface
temperature is set to 130 °C, for 45 minutes, rolled and cooled,
and is pulverized by a pulverizer to obtain a master batch pigment.

30 [0214]

Polyol resin 1 100 parts

Master batch 8 parts

Charge control agent (BONTRON E-84,

available from Orient Chemical Industries, Ltd.) 2 parts
Wax (a fatty acid ester wax, melting point: 83 °C,
viscosity: 280 mPa · s (90 °C)) 5 parts

The above material is mixed in a mixer, is then melted and kneaded
5 in a two-roll mill for three or more times, and the kneaded material
is rolled and cooled. The resulting material is pulverized in a
pulverizer (I-Type Mill : available from Nippon Pneumatic MFG. Co.,
Ltd.) of a jet mill breaker disc system, and subjected to air
classification by action of a revolving current (DS classifier :
10 available from Nippon Pneumatic MFG. Co., Ltd.) and thus yellow
particles having a volume-average particle diameter of 5.5 μm is
obtained. The yellow particles are further mixed with 3.0% by
weight of the oxide fine particles 1 and 1.0% by weight of a
hydrophobic silica (HDK H 2000, available from Clariant Japan K.K.)
15 having a primary particle diameter of 10 nm in a Henschel mixer,
the resulting mixture is passed through a sieve with an aperture
of 50 μm to remove aggregates. The yellow toner 1 is thus obtained.
A dispersion diameter of the wax in the toner is 0.5 μm.

[0215]

20 Magenta toner

Water	600 parts
Pigment red 57 hydrous cake (solid content 50%)	1200 parts
Polyol resin 1	1200 parts

The above materials are mixed in a Henschel mixer and a mixture
25 in which pigment aggregates are impregnated with water is obtained.
The mixture is kneaded in a twin-roll mill, of which a roll surface
temperature is set to 130 °C, for 45 minutes, rolled and cooled,
and is pulverized by a pulverizer to obtain a master batch pigment.

[0216]

30 Polyol resin 1	100 parts
Master batch	8 parts
Charge control agent (BONTRON E-84, available from Orient Chemical Industries, Ltd.)	2 parts

Wax (a fatty acid ester wax, melting point: 83 °C,
viscosity: 280 mPa · s (90 °C)) 5 parts

The above material is mixed in a mixer, is then melted and kneaded in a two-roll mill for three or more times, and the kneaded material is rolled and cooled. The resulting material is pulverized in a pulverizer (I-Type Mill : available from Nippon Pneumatic MFG. Co., Ltd.) of a jet mill breaker disc system, and subjected to air classification by action of a revolving current (DS classifier : available from Nippon Pneumatic MFG. Co., Ltd.) and thus magenta particles having a volume-average particle diameter of 5.5 μm is obtained. The magenta particles are further mixed with 3.0% by weight of the oxide fine particles 1 and 1.0% by weight of a hydrophobic silica (HDK H 2000, available from Clariant Japan K.K.) having a primary particle diameter of 10 nm in a Henschel mixer, the resulting mixture is passed through a sieve with an aperture of 50 μm to remove aggregates. The magenta toner 1 is thus obtained. A dispersion diameter of the wax in the toner is 0.5 μm .

[0217]

Cyan toner

20	Water	600 parts
	Pigment Blue 15:3 hydrous cake (solid content 50%)	1200 parts
	Polyol resin 1	1200 parts

The above materials are mixed in a Henschel mixer and a mixture in which pigment aggregates are impregnated with water is obtained. The mixture is kneaded in a twin-roll mill, of which a roll surface temperature is set to 130 °C, for 45 minutes, rolled and cooled, and is pulverized by a pulverizer to obtain a master batch pigment.

[0218]

	Polyol resin 1	100 parts
30	Master batch	8 parts
	Charge control agent (BONTRON E-84, available from Orient Chemical Industries, Ltd.)	2 parts

Wax (a fatty acid ester wax, melting point: 83 °C,
viscosity: 280 mPa · s (90 °C)) 5 parts

The above material is mixed in a mixer, is then melted and kneaded in a two-roll mill for three or more times, and the kneaded material is rolled and cooled. The resulting material is pulverized in a pulverizer (I-Type Mill : available from Nippon Pneumatic MFG. Co., Ltd.) of a jet mill breaker disc system, and subjected to air classification by action of a revolving current (DS classifier : available from Nippon Pneumatic MFG. Co., Ltd.) and thus cyan particles having a volume-average particle diameter of 5.5 μm is obtained. The cyan particles are further mixed with 3.0% by weight of the oxide fine particles 1 and 1.0% by weight of a hydrophobic silica (HDK H 2000, available from Clariant Japan K.K.) having a primary particle diameter of 10 nm in a Henschel mixer, the resulting mixture is passed through a sieve with an aperture of 50 μm to remove aggregates. The cyan toner 1 is thus obtained. A dispersion diameter of the wax in the toner is 0.5 μm.

[0219]

Embodiment 2 to 7

Toner and a developer are produced and evaluated in a same method as in embodiment 1, except that oxide fine particles 2 to 7 in Table 2 are used.

[0220]

Embodiment 8

An evaluation is carried out in a same method as in embodiment 1, except that the resin is changed to a polyester resin (prepared from fumaric acid, polyoxypropylene - (2.2) - 2, 2 - bis (4 - hydroxyphenyl) propane, polyoxyethylene - (2.3) - 2,2 - bis (4 - hydroxyphenyl) propane, and trimellitic anhydride, wherein an acid value: 10, a hydroxyl value: 30, Mn: 5000, Mw/Mn: 10, Mp: 9000, Tg: 61 °C, and a softening point: 108 °C).

[0221]

Embodiment 9

An evaluation is carried out in a same method as in Embodiment 1, except that a test machine B is used.

[0222]

Embodiment 10

5 An evaluation is carried out in a same method as in Embodiment 1, except that a test machine C is used.

[0223]

Embodiment 11

10 An evaluation is carried out in a same method as in Embodiment 1, except that a test machine D is used.

[0224]

Embodiment 12

15 An evaluation is carried out in a same method as in Embodiment 1, except that toner is produced without adding wax and a test machine E is used.

[0225]

Comparative example 1 to 4

20 Toner and a developer are produced and evaluated in a same method as in embodiment 1, except that oxide particles 8 to 11 in Table 2 are used.

[0226]

[Effects of the Invention]

25 It is found that by using an external additive for electrophotographic toner, characterized in that the external additive is formed of an oxide fine particle including at least a silicon element, the oxide fine particle has a primary particle diameter R of 30 to 300 nm, a standard deviation δ of a particle size distribution of R is $R/4 \leq \delta \leq R$, and a shape of the external additive is substantially spherical with circularity SF1 and
30 circularity SF2 of 100 to 130 and 100 to 125, respectively, a high quality image may be formed so that even after toner is stored under a high temperature/humidity environment, the external additive may not be buried in the toner and may sufficiently exhibit functions

as a fluidizing and a charge auxiliary agent, even after the toner is stored under a low temperature/humidity environment, an image with stable quality may be provided by preventing a charge characteristic from abnormally increasing, and furthermore, the image is excellent in an aggregation characteristic of the toner during transfer and compression, and transfer and development characteristic in which adhesion force among toner particles after the toner is subjected to stress in development device is appropriately controlled.

[0227]

Further, by improving the transfer characteristic of toner, following things are achieved; prevention of an abnormal image such as a hollow defect in a character part from occurring, reduction of an amount of waste toner by improving a toner transfer rate, reduction of an amount of toner consumption, uniformity in a solid image by improving supply characteristic of toner, reduction of transfer dust, and so forth, improvement of thin line reproducibility, reduction of background soil by improving environmental stability of charging under a high temperature/humidity or a low temperature/humidity environment, and prevention of toner dispersion. Further, a printed material with excellent thermal storage stability, color reproducibility, clearness, glossiness, transparency, and a fixing characteristic may be obtained.

[0228]

[Table 1]

Table 1: Evaluated result

	Test machine	External additive embedding	Toner dispersion	Hollow defects	Toner transfer rate	Toner supply	Transfer dust	Thin line reproducibility	Background soil
Em. 1	A	○	○	○	○	○	○	○	○
Em. 2	A	△	○	○	○	○	○	○	○
Em. 3	A	◎	○	○	○	○	○	○	○
Em. 4	A	◎	○	○	○	○	○	○	○

Em. 5	A	○	○	○	○	○	○	○	○
Em. 6	A	○	○	○	○	○	○	○	○
Em. 7	A	○	○	◎	◎	△	◎	◎	○
Em. 8	A	◎	○	○	◎	○	○	○	○
Em. 9	B	○	○	○	○	○	△	○	○
Em. 10	C	○	△	○	△	○	△	○	○
Em. 11	D	○	○	○	○	○	△	○	○
Em. 12	E	○	○	○	○	◎	○	○	○
Comp. Ex. 1	A	◎	×	×	×	×	×	△	×
Comp. Ex. 2	A	△	×	×	×	○	×	△	×
Comp. Ex. 3	A	×	×	×	○	○	○	×	○
Comp. Ex. 4	A	○	×	×	×	×	×	△	×

	Image density	Heat resistance	Transparency	Clearness	Color reproduci- bility	Glossiness	Charge stability at high temp. & humidity	Charge stability at low temp. & humidity	Fixing character- istic
Em. 1	◎	○	○	○	◎	◎	○	○	○
Em. 2	◎	○	○	○	◎	◎	○	○	○
Em. 3	◎	○	○	○	◎	◎	△	△	○
Em. 4	◎	○	○	○	◎	◎	○	◎	○
Em. 5	◎	○	○	○	◎	◎	◎	◎	○
Em. 6	◎	△	○	○	◎	◎	◎	◎	○
Em. 7	◎	△	○	○	◎	◎	○	○	○
Em. 8	◎	◎	◎	◎	◎	◎	○	○	◎
Em. 9	◎	○	○	○	○	◎	○	○	○
Em. 10	◎	○	○	○	○	◎	△	△	○
Em. 11	◎	○	○	○	○	◎	○	○	○
Em. 12	◎	○	○	○	○	◎	○	○	○
Comp. Ex. 1	△	○	△	○	○	△	×	×	×

Comp. Ex. 2	△	△	△	○	○	○	×	×	○
Comp. Ex. 3	◎	○	○	○	◎	◎	○	○	○
Comp. Ex. 4	×	△	△	○	○	○	×	×	○

[0229]

Table 2 Production examples of oxide fine particles and embodiments

	Oxide fine particle	Primary particle diameter (nm)	Standard deviation of primary particle diameter (nm)	Circularity SF1	Circularity SF2	Dispersion of elements
Em.1	Oxide fine particle 1	110	50	120	109	Even
Em.2	Oxide fine particle 2	80	70	125	121	Even
Em.3	Oxide fine particle 3	170	60	128	110	Uneven
Em.4	Oxide fine particle 4	210	70	115	117	Even
Em.5	Oxide fine particle 5	120	40	118	106	Even
Em.6	Oxide fine particle 6	150	75	119	110	Even
Em.7	Oxide fine particle 7	140	50	121	123	Even
Comp. Ex.1	Oxide fine particle 8	310	150	122	110	Even
Comp. Ex.2	Oxide fine particle 9	28	10	121	121	Even
Comp. Ex.3	Oxide fine particle 10	120	11	118	110	Even
Comp. Ex.4	Oxide fine particle 11	121	65	131	127	Even

5 [Brief Description of the Drawings]

[Fig. 1]

Fig. 1 is a schematic drawing illustrating one embodiment in which toner and a developer according to the present invention are used.

[Fig. 2]

10 Fig. 2 is a schematic drawing illustrating one embodiment in which the toner and the developer according to the present invention are used.

[Fig. 3]

15 Fig. 3 is a schematic drawing illustrating one embodiment in which the toner and the developer according to the present invention are used.

[Fig. 4]

Fig. 4 is a schematic drawing illustrating one embodiment in which the toner and the developer according to the present invention are used.

5 [Fig. 5]

Fig. 5 is a schematic drawing illustrating one embodiment in which the toner and the developer according to the present invention are used.

[Fig. 6]

10 Fig. 6 is a schematic drawing illustrating one embodiment in which the toner and the developer according to the present invention are used.

[Reference Numerals]

10	Photoconductor
15	20 Charging roller
	30 Exposure device
	40 Development apparatus
	41 Development belt
	42 Development tank
20	43 Scoop roller
	44 Application roller
	45 Development unit
	50 Intermediate transfer body
	51 Suspension roller
25	52 Corona charging device
	53 Constant current source
	60 Cleaning device
	70 Discharge lamp
	80 Transfer roller
30	90 Cleaning device
	100 Transfer paper

[Selected Drawing] None

[Name of Document] Abstract of the Disclosure

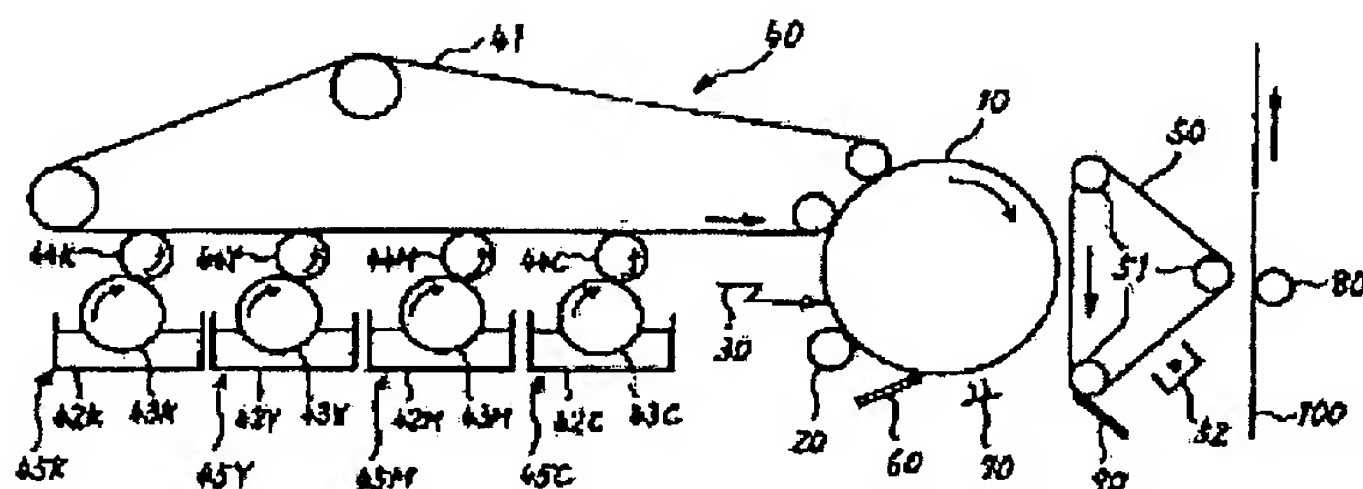
[Abstract]

[Objectives of the Invention] To provide an external additive for
5 electrophotography in which the external additive may not be buried
in toner and may satisfactorily exert functions as a fluidizing and
a charge auxiliary agent even when the toner is subjected to
agitation and charging after being stored under a high
temperature/humidity environment, and stable image quality may be
10 achieved by preventing charge from abnormally increasing even after
the toner is stored in a low temperature/humidity environment,
electrophotographic toner and an electrophotographic developer
including the external additive for electrophotography.

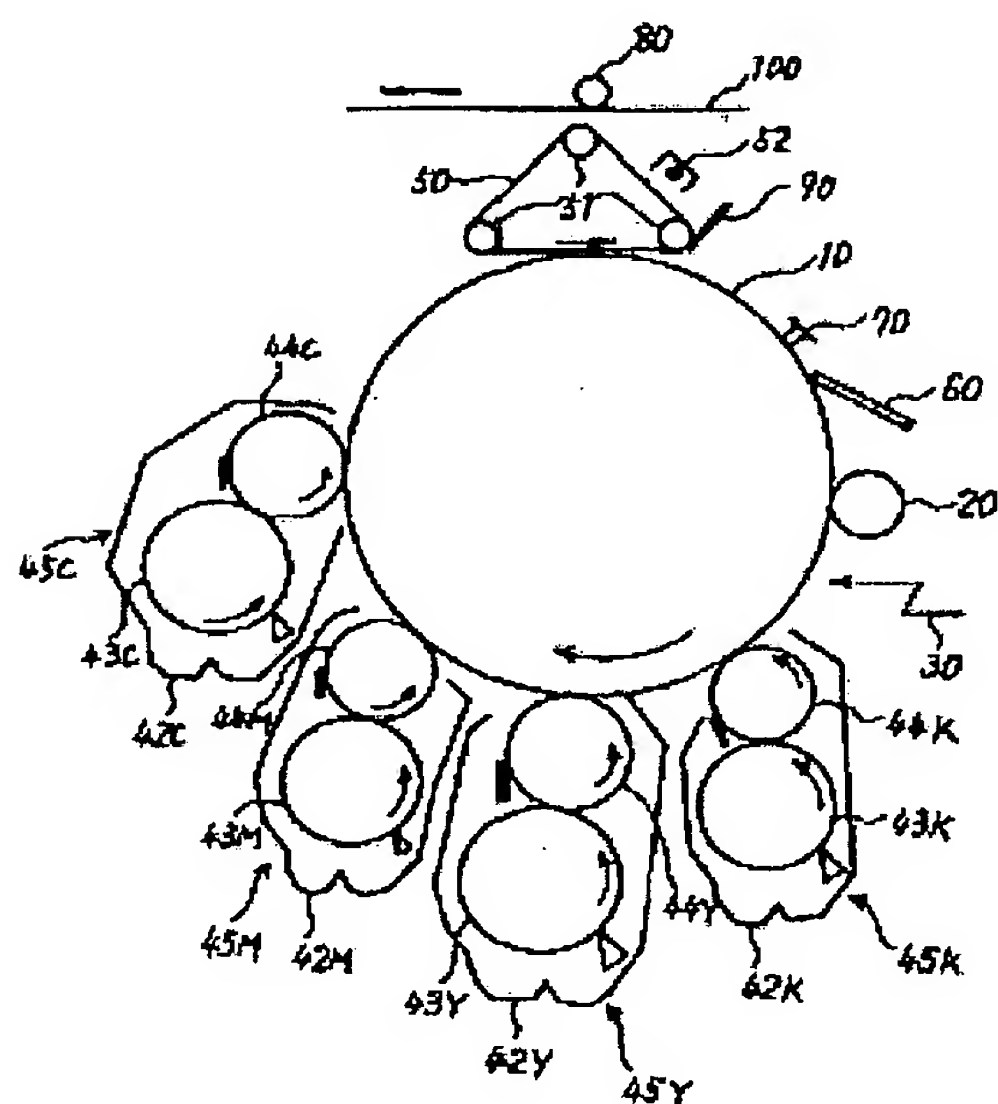
[Means for Achieving the Objectives] An external additive for
15 electrophotographic toner, characterized in that the external
additive is formed of oxide fine particles including at least a
silicon element, the oxide fine particle has a primary particle
diameter R (number average) of 30 to 300 nm, a standard deviation
 δ of a particle size distribution of R is $R/4 \leq \delta \leq R$, and a shape
20 of the external additive is substantially spherical with
circularity SF1 and circularity SF2 of 100 to 130 and 100 to 125,
respectively.

[Name of Document]

[Fig. 1]

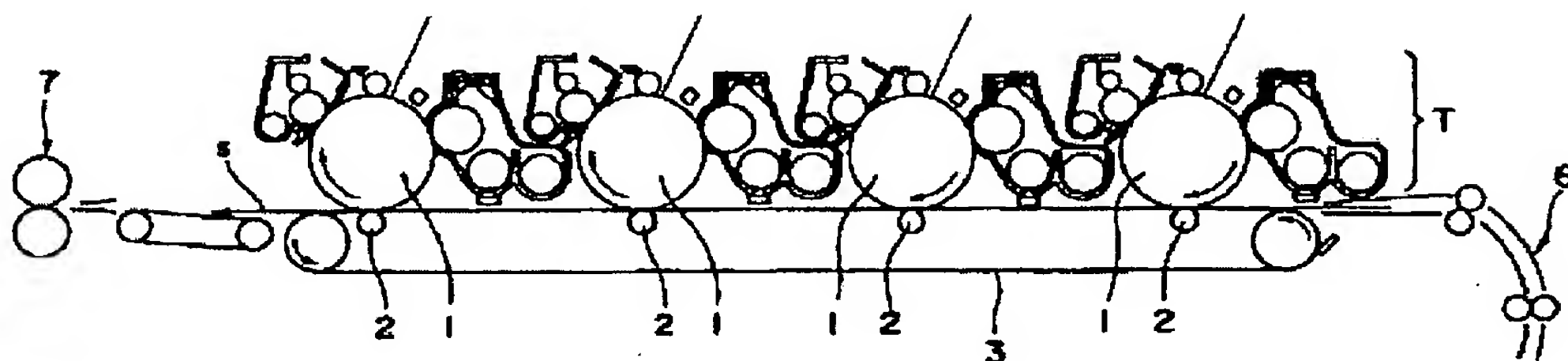


[Fig. 2]

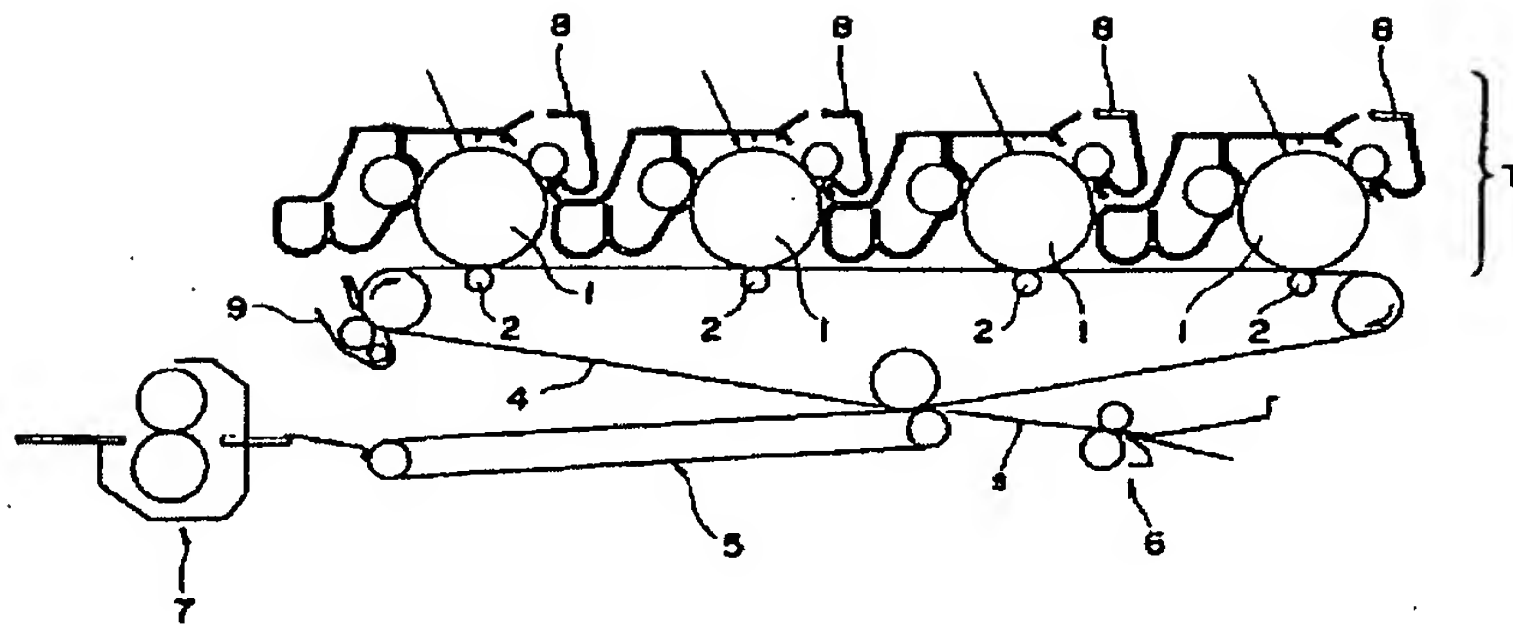


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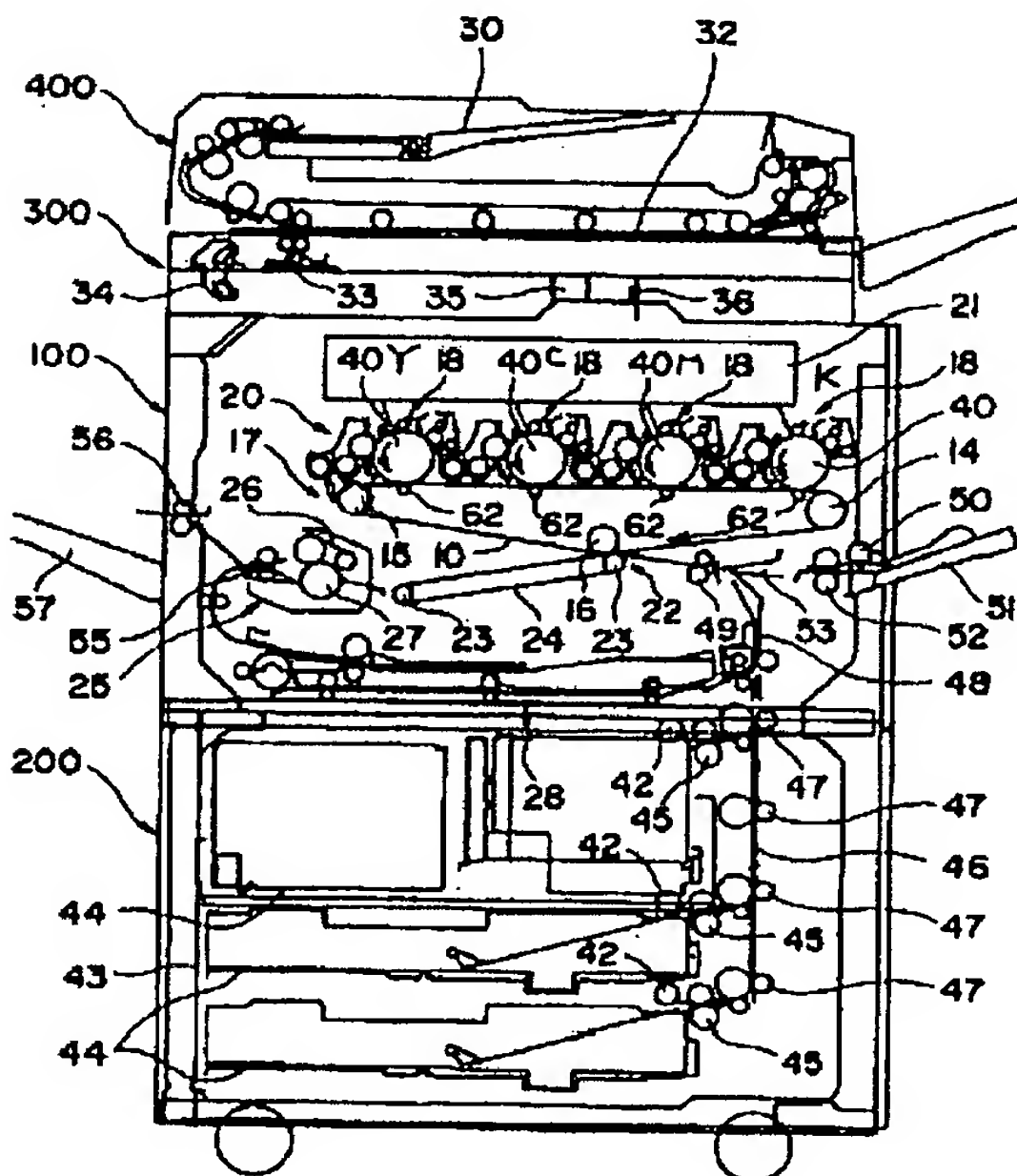
[Fig. 3]



[Fig. 4]



[Fig. 5]



5 [Fig. 6]

